ORGIA INSTITUTE OF TECHNOLOGY OFFICE OF CONTRACT ADMINISTRATION

PROJECT ADMINISTRATION DATA SHEET

[x] ORIGINAL [ ] REVISION NO. _____

ject No. E-19-660 DATE 8/27/82

ject Director: Peter Skelland School/~~Lab~~ ChE

nsor: National Science Foundation
Washington, DC 20550

e Agreement: Grant No. CPE-8203872

ard Period: From 8/1/82 To ~~1/31/85~~ 6-30-86 (Performance) 4/30/85 (Reports)

nsor Amount: $125,672 Contracted through:

t Sharing: $ 3,918 (E-19-327) GTRI/~~GIT~~

le: Disperison and Mass Transfer in Agitated Liquid-Liquid Systems

MINISTRATIVE DATA OCA Contact Linda H. Bowman x4820

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fense Priority Rating: NA Security Classification: NA

STRICTIONS

Attached NSF Supplemental Information Sheet for Additional Requirements.

vel: Foreign travel must have prior approval – Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of $500 or 125% of approved proposal budget category.

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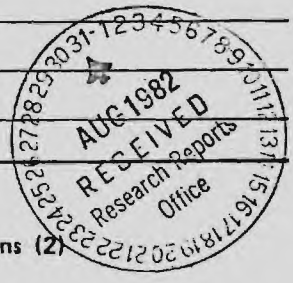

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EORGIA INSTITUTE OF TECHNOLOGY OFFICE OF CONTRACT ADMINISTRATION

SPONSORED PROJECT TERMINATION/CLOSEOUT SHEET

Date 2/27/87

oject No. E-19-660 School/~~Lab~~ CHE

cludes Subproject No.(s) N/A

oject Director(s) Anthony Skelland GTRC /~~GIT~~

onsor National Science Foundation, Washington, DC 20550

le Disperison and Mass Transfer in Angitated Liquid-Liquid Systems

fective Completion Date: 6/30/86 (Performance) 9/30/86 (Reports)

ant/Contract Closeout Actions Remaining:

- [X] None
- [ ] Final Invoice or Final Fiscal Report
- [ ] Closing Documents
- [ ] Final Report of Inventions
- [ ] Govt. Property Inventory & Related Certificate
- [ ] Classified Material Certificate
- [ ] Other

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Angela Jones
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APPENDIX VII

NATIONAL SCIENCE FOUNDATION
Washington, D.C. 20550

FINAL PROJECT REPORT
NSF FORM 98A

PLEASE READ INSTRUCTIONS ON REVERSE BEFORE COMPLETING

PART I–PROJECT IDENTIFICATION INFORMATION

1. Institution and Address	2. NSF Program	3. NSF Award Number
Georgia Institute of Technology Atlanta, GA 30332	Separation & Purification	CPE82-03872
	4. Award Period From 8/1/82 To 6/31/86	5. Cumulative Award Amount $199,182

6. Project Title
Dispersion and Mass Transfer in Agitated Liquid-Liquid Systems.

PART II–SUMMARY OF COMPLETED PROJECT *(FOR PUBLIC USE)*

The grant partially supported a three-part program of experimental work.

Part I comprised a visual determination and correlation of the minimum impeller speeds needed for complete dispersion of one immiscible liquid in another in baffled vessels. Variables included impeller type, location, and number, vessel size, physical properties, and volume fraction of disperse phase. Data from 481 runs from three different sources were correlated with an average absolute deviation of 12.7%. The correlation was then extended to incorporate the effects of surface active contamination - a common industrial occurrence.

In Part II, 180 experimental runs were performed to measure, interpret, and correlate the continuous phase mass transfer coefficient k_c in agitated liquid-liquid systems. It was found that k_c was related to impeller speed, viscosity and diffusivity as $k_c \propto N^{1.5} \mu_c^{-1/3} D^{2/3}$. The corresponding disperse phase mass transfer coefficient k_d was measured in 86 runs on three systems. The results were used to select between two provisional theoretical models, and were also empirically correlated.

Part III explored the effects of interaction between adjacent droplets on mass transfer, and the effects of surface active contamination on jet breakup, drop formation, and drop oscillation.

The results should facilitate design of extraction columns, mixer-settlers, batch polymerizers, and chemical reactors.

PART III–TECHNICAL INFORMATION *(FOR PROGRAM MANAGEMENT USES)*

1. ITEM *(Check appropriate blocks)*	NONE	ATTACHED	PREVIOUSLY FURNISHED	TO BE FURNISHED SEPARATELY TO PROGRAM	
				Check (✓)	Approx. Date
a. Abstracts of Theses		X(5)			
b. Publication Citations		X(8)			
c. Data on Scientific Collaborators		X(8)			
d. Information on Inventions	X				
e. Technical Description of Project and Results		X			
f. Other *(specify)*					

2. Principal Investigator/Project Director Name *(Typed)*	3. Principal Investigator/Project Director Signature	4. Date
A. H. P. Skelland		2/17/87

NSF Form 98A (3-83) Supersedes All Previous Editions

Form Approved OMB No. 3145-0058

Title pages, Abstract/Summary and Acknowledgement pages from Ph.D. and M.S. theses in Chemical Engineering at the Georgia Institute of Technology based on work partially supported by NSF grants numbered CP82-03872 and CPE82-03872/01.

THE DEPENDENCE OF THE CONTINUOUS PHASE MASS TRANSFER COEFFICIENTS ON MOLECULAR DIFFUSIVITY FOR LIQUID-LIQUID EXTRACTION IN AGITATED VESSELS

A THESIS
Presented to
The Faculty of the Division of Graduate Studies

By

Lebone Tiisang Moeti

In Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy in Chemical Engineering

Georgia Institute of Technology
October, 1986

THE DEPENDENCE OF THE CONTINUOUS PHASE
MASS TRANSFER COEFFICIENTS ON MOLECULAR DIFFUSIVITY FOR
LIQUID-LIQUID EXTRACTION IN AGITATED VESSELS

Approved:

A. [illegible], Chairman

Larry J. [illegible]

Charles W. Gorton

Russell G. Heikes

Michael J. Matteson

Date Approved by Chairman 11/6/86

ACKNOWLEDGEMENTS

The author wishes to express his sincere gratitude and appreciation to Dr. A.H.P. Skelland for his guidance and encouragement throughout the course of this study. He also wishes to thank Dr. C.L. Liotta of the Chemistry Department for his help in clarifying the interactions between sucrose and acids.

Financial support from National Science Foundation grant numbers CPE 82-03872 and CPE 82-03872/01 is gratefully acknowledged.

SUMMARY

This research examined the influence of the diffusivity on the continuous phase mass transfer coefficient to identify the mechanism of mass transfer for liquid-liquid extraction in agitated vessels. By identifying the prevailing mechanism of mass transfer, optimized design, future mathematical modeling, and scale-up considerations can be improved for applications such as mixer-settler design, emulsion polymerization, and reactor design.

Results of this study are summarized below:

1. For liquid-liquid extraction in agitated vessels k_c has been shown to be proportional to $D^{2/3}$. The exponent on the diffusivity was determined by using temperature variation to increase the range of D. With a 7.8-fold range of D, exponents of 2/3 on the diffusivity were found for both high and low interfacial tension systems, in agreement with the exponent on diffusivity at each of 10 temperatures in runs where only D varied (approx. 2-fold). Therefore, mass transfer mechanisms that invoke $k_c \propto D^{2/3}$ are appropiate for liquid-liquid extraction in agitated vessels. Exponents on D of 2/3 indicate that drops in agitated vessels behave like rigid spheres with no internal circulation. However, the phenom-

ena of drop breakage, which result in the formation of new and uncontaminated surface and in surface deformation, is felt to be the key difference between solid-liquid ($k_c \propto D^{2/3}$) and liquid-liquid mass transfer. The majority of solid-liquid mass transfer studies that report $k_c \propto D^{1/2}$ have involved the use of sucrose solutions for viscosity variation, which undergo acidic hydrolysis and hydrogen bonding in the presence of acids. These interactions between sucrose and acids in aqueous solution result in reduced mobility of acid solutes and lead to incorrect conclusions regarding the effect of D on k_c.

2. The exponent on the impeller speed, N, is consistently higher for liquid-liquid mass transfer than for solid-liquid mass transfer, despite the fact that both disperse phases behave as rigid spheres ($k_c \propto D^{2/3}$); this is probably due to the drop breakage effect in liquid-liquid systems. For solid-liquid systems which do not undergo particle breakage, the exponents on N are thus lower.

3. The effect of the continuous phase viscosity, μ_c, on the mass transfer coefficient, k_c, was found to be smaller ($k_c \propto \mu_c^{1/3}$) in this research than in studies where sucrose was used to increase viscosity. The interaction between sucrose and acids, as described earlier, resulted in higher exponents on viscosity for investigators who attempted to increase the range of the Schmidt number by using sucrose.

4. A theory has been developed to describe the process of

mass transfer that occurs in an agitated vessel. The Film-Penetration Theory with Periodically Varying Rate of Surface Renewal allows for the different degrees of turbulence that exist at various locations within the agitated vessel.

5. Semi-theoretical dimensionless correlations for the mass transfer coefficient, k_c, have been developed based on the theory of isotropic turbulence. The primary correlation involving the effect of drop breakage is given by

$$\frac{k_c d_p}{D} = 25.459\left(\frac{\mu_c}{\rho_c D}\right)^{1/3}\left(\frac{d_I^{\,2} N \rho_c}{\mu_c}\right)^{2/3}\left(\frac{d_p}{d_I}\right)^{2/9}\left(\frac{d_p}{T}\right)^{2/3} \cdot \left(\frac{d_p^{\,3} N^2 \rho_d}{\sigma}\right)^{5/12} \phi^{-1/2}$$

with an overall absolute deviation of 28.13% for the systems of this research. A secondary correlation was developed so that k_c would be independent of the drop diameter, d_p, since the effect of d_p on k_c cannot be determined experimentally for liquid-liquid systems. The correlation based on the contamination of the drop surface, such that k_c is independent of d_p, is as follows:

$$\frac{k_c d_p}{D} = 1.237(10^{-5})\left(\frac{\mu_c}{\rho_c D}\right)^{1/3}\left(\frac{d_I^2 N \rho_c}{\mu_c}\right)^{2/3}\left(\frac{d_I N^2}{g}\right)^{5/12}\left(\frac{d_I}{d_p}\right)^2\left(\frac{d_p}{T}\right)^{1/2} \cdot \left(\frac{\rho_d d_p^2 g}{\sigma}\right)^{5/4} \phi^{-1/2}$$

for the three systems used in this study. The secondary correlation had an overall absolute deviation of 19.71%. Although the secondary correlation, involving drop surface contamination, had a lower deviation than the correlation involving drop breakage, the secondary correlation did not fit continuous phase mass transfer data taken from other investigators as well as the correlation involving drop breakage. Therefore the correlation based on drop breakage was chosen as the general correlation of k_c. In addition, the positive directional effect of d_p on k_c was supported by the development based on the breakage frequency function (Equation 5-33).

6. The Sauter-mean droplet diameters predicted by the correlations of Hong and Lee (1985) and Skelland and Hu (1986), which are perhaps the only correlations for unsteady-state drop sizes in agitated vessels, were compared to the drop sizes of the present research and showed an overall absolute deviation of 45.2% for both correlations.

EFFECTS OF SURFACE ACTIVE AGENTS ON MINIMUM IMPELLER SPEEDS FOR LIQUID-LIQUID DISPERSION IN BAFFLED VESSELS

A THESIS

Presented to
The Faculty of the Division of Graduate Studies
by

Lebone Tiisang Moeti

In Partial Fulfillment
of the Requirements for the Degree
Master of Science in Chemical Engineering

Georgia Institute of Technology

January, 1984

EFFECTS OF SURFACE ACTIVE AGENTS ON
MINIMUM IMPELLER SPEEDS FOR
LIQUID-LIQUID DISPERSION IN BAFFLED
VESSELS

Approved:

A. H. Peter Skelland, Chairman

Michael J. Matteson

Allan S. Myerson

Date Approved by Chairman Feb 20, 1984

ACKNOWLEDGEMENTS

The author wishes to express his sincere gratitude and appreciation to Dr. A. H. P. Skelland for his guidance and encouragement throughout the course of this study.

Financial support from National Science Foundation, Grant No. CPE 82-03872, is gratefully acknowledged.

SUMMARY

In industrial operations, the presence of surface active contaminants in trace amounts is widely encountered and the effects of these materials in distorting performance from that predicted by studies with pure systems need to be accommodated. In the present study, the effects of nonionic, anionic, and cationic surface active agents (SAA) on the minimum impeller speed for complete dispersion in agitated vessels were examined. The Skelland and Ramsay (1983) correlation

$$\frac{N_{min} D^{1/2}}{g^{1/2}} = C \left(\frac{T}{D}\right)^{\alpha} \left(\frac{\mu_M}{D^{3/2} \rho_M g^{1/2}}\right)^{0.09} \left(\frac{\Delta\rho}{\rho_M}\right)^{0.42} \left(\frac{\sigma}{D^2 \rho_M g}\right)^{0.04} \phi^{0.05}$$

worked well in the prediction of minimum impeller speed using diminished interfacial tensions due to SAA, with an overall deviation of 11.67%. In contrast to some droplet phenomena (Mekasut, et. al., 1979), the difference between deviations among the surfactant systems was accounted for by experimental error and verified by two methods of statistical analysis.

AGITATION FOR DISPERSION AND SURFACTANT EFFECTS ON DROPLET BEHAVIOR IN LIQUID-LIQUID SYSTEMS

A THESIS

Presented to

The Faculty of the Division of Graduate Studies

By

George Ramsay

In Partial Fulfillment

of the Requirements for the Degree

Master of Science in Chemical Engineering

Georgia Institute of Technology

August 1984

AGITATION FOR DISPERSION AND SURFACTANT EFFECTS ON DROPLET BEHAVIOR IN LIQUID-LIQUID SYSTEMS

Approved:

A. H. P. Skelland, Chairman

Michael J. Matteson

Larry C. Forney

ACKNOWLEDGEMENTS

The author wishes to express his sincere gratitude and appreciation to Dr. A. H. P. Skelland for making this thesis possible, to Dr. Michael J. Matteson for his guidance and for being on my Reading Committee, to Dr. Gary Poehlein and Dr. Jude T. Sommerfeld for their patience and understanding, and to Dr. Larry J. Forney for being on my Reading Committee.

This work was partially supported by National Science Foundation Grant Numbers CPE 80-19617 and CPE 82-03872.

ABSTRACT

Data from three different sources have been pooled to obtain an empirical correlation of the minimum agitator speed needed to obtain complete liquid-liquid dispersion in baffled vessels. The variables include five common types of impeller, two generating axial flow and three radial flow, in four locations; fluid properties in eleven systems; tank diameter; liquid height; and volume fraction of the disperse phase.

Observations from 481 runs were correlated with an average absolute deviation of 12.7% by the expression

$$(N_{Fr})_{min} = C^2 \left(\frac{T}{D}\right)^{2\alpha} \phi^{0.106} (N_{Ga} N_{Bo})^{-0.084}$$

where C and α are tabulated according to impeller type, location, and -- in some cases -- the ratio H/T. Scale-up relationships are developed from the above expression.

Drop size, terminal velocity, and the onset of oscillation of chlorobenzene drops falling through water were determined in systems containing anionic, cationic, or nonionic surfactants. Surfactant concentration, nozzle diameter, and nozzle velocity were varied to cover the range of most industrial applications of drop formation from circular orifices in the nonjetting region.

Existing correlations for drop size and terminal velocity in uncontaminated systems are shown to be adequate in the presence of surfactants (SAA) when used with the diminished value of interfacial tension due to the SAA. The applicability of criteria for the onset of oscillation in pure systems to those containing surfactants is examined.

THE EFFECTS OF SURFACE-ACTIVE AGENTS ON JET BREAK-UP IN NEWTONIAN LIQUID-LIQUID SYSTEMS

A THESIS

Presented to

The Faculty of the Division of Graduate Studies

By

Perry George Walker

In Partial Fulfillment

of the Requirements for the Degree

Master of Science in Chemical Engineering

Georgia Institute of Technology

August, 1984

THE EFFECTS OF SURFACE-ACTIVE AGENTS ON JET BREAK-UP IN NEWTONIAN LIQUID-LIQUID SYSTEMS

Approved:

A. H. Peter Skelland,
Chairman

Michael J. Matteson

Larry J. Forney

Date approved by Chairman

ABSTRACT OF THESIS

It has been found that drop formation under jetting conditions is preferable in liquid-liquid extraction in perforated-plate columns. Studies have been made to predict the size of drops resulting from jet break-up in Newtonian liquid-liquid systems.

Because many industrial applications of liquid-liquid extraction are carried out in the presence of surface-active contaminants, it is important to study their effects on the process. This work examines the effect of surfactants on jet length, jet contraction, and drop size from jet break-up in Newtonian liquid-liquid systems.

A total of 143 runs were made using the methods of Johnson (10) and Huang (9), and 501 photographs were taken resulting in data for 143 jets and 3026 drops. These data were used to test the adequacy of the correlations of Skelland and Huang (29) in the presence of surface-active agents.

Results of the study are summarized below:

Jet Length

Using the Skelland and Huang (29) correlation

$$L_j/d_n = 5.0767\ (\Delta N_{We})^{0.5499}(\mu_c/\mu_d)^{0.5245}$$

with the diminished values of interfacial tension due to the surfactant, resulted in a mean absolute error of 38.10% with standard deviation of 34.26. Skelland and Huang (29) reported an absolute deviation of 39.69% with their data. A regression analysis and a one way analysis of variance were performed on these data, showing good agreement between the values of jet length predicted by the previous correlation and the present data provided that the system is one of high interfacial tension.

Drop Size

Plots of U_o/U_{om} vs. d_D/d_{jm} were made for all runs as suggested by Huang (9) and Johnson (10). Comparison of the data with the values predicted by the Skelland and Huang (29) correlation

$$d_D/d_{jm} = 3.0704 - 0.1701\ (U_o/U_{om}) + 0.0487\ (U_o/U_{om})^2$$

using diminished values of interfacial tension due to the presence of surfactants, showed a mean absolute error of 15.73%. Most of the experimental values were lower than those predicted. A better fit was obtained using Skelland and Huang's correlation for very low mass transfer rates

$$d_D/d_{jm} = 2.7270 - 0.2028\ (U_o/U_{om}) + 0.0731\ (U_o/U_{om})^2$$

(11.08% error).

The greater dependence on flow rate exhibited by this correlation makes it more flexible, and will thus show more accuracy for various values of interfacial tension. For best results, however, one should use a separate correlation for each liquid-liquid system and each relative nozzle diameter.

Jet Contraction

Christiansen and Hixson (3), Johnson (10), and Huang (9) used a plot of d_n/d_{jc} vs. d_n/ℓ_s to derive a least-squares line for their data. Skelland and Huang (29) reported the following correlation

$$d_n/d_{jc} = 2.7350 \left(d_n/\ell_s \right) + 0.5718$$

from their data, which when used with the results of the experiments and diminished values of interfacial tension due to the presence of surfactants gave a mean absolute error of 13.03%. Predicted values were higher than the experimental values. This was probably due to a reduction in the rate of mass transfer caused by the surfactants. Because of the high probability of experimental error, one can ignore the small amount of error caused by the reduction in mass transfer rates. Two forms of statistical analysis were used to verify that this correlation is relatively accurate in the presence of surfactants using the reduced values for interfacial tension caused by the surfactants.

EFFECTS OF SURFACE ACTIVE AGENTS ON DROP SIZE IN LIQUID-LIQUID SYSTEMS

A THESIS

Presented to

The Faculty of the Division of Graduate Studies

By

Elizabeth Ann Slaymaker

In Partial Fulfillment

of the Requirements for the Degree of

Master of Science in Chemical Engineering

Georgia Institute of Technology

December, 1985

SUMMARY

The interfacial area available for mass transfer is an important factor in liquid-liquid extraction and other separation processes; thus, knowledge about the size of drops formed at perforations or nozzles is fundamental.

Most of the previous work on drop size has been for pure liquid-liquid systems with special care taken to ensure absence of contaminants. In practice, however, almost all systems are plagued with the presence of surface active impurities.

The purpose of this study was to attempt to determine whether the effect of such surface active impurities is simple or complex. Drop size data from systems containing surfactants was compared to the values obtained from the Scheele and Meister correlation for drop size using reduced surface tensions. Only the effect of surfactants on surface tension was considered. For pure systems, the Scheele and Meister correlation is considered to be "the best compromise between accuracy and ease of use."

With the simplifying assumption that the only effect of the addition of surfactants to a system was on the interfacial tension, use of the Scheele and Meister correlation for contaminated systems as well as pure systems appears to be justified. It is important to note, however, that systems studied were chosen so that mass transfer effects were negligible. The complicated nature of the effects of surfactants on mass transfer may lead to the conclusion that the Scheele and Meister

correlation cannot be applied to contaminated systems in which mass transfer occurs. This is a possible subject for future work.

Papers either published or in the course of publication, based on work partially supported by NSF grants numbered CPE82-03872 and CPE82-03872/01

Effects of Interaction Between Circulating or Oscillating Droplets on Drop Formation, Free Fall, and Mass Transfer

A. H. P. SKELLAND and NICHOLAS C. VASTI[1]

School of Chemical Engineering, The Georgia Institute of Technology, Atlanta, GA 30332, USA

The effects of interaction between circulating or oscillating droplets on drop formation, terminal velocity, and the continuous phase mass transfer coefficient during free fall were studied. To examine the mass transfer coefficient during free fall both end effects were eliminated by subtracting the mass transferred in a short column from that transferred in a long column. A correlation for the prediction of drop volume based on the equations of Scheele and Meister (1968) for non-jetting drops, and of Meister and Scheele (1969a, 1969b) for drops formed under jetting conditions, has been developed. The terminal velocity of the drops was enhanced by vertical interaction with adjacent drops at values of $L/d_p < 5$. The continuous phase mass transfer coefficient for circulating drops was reduced due to vertical interaction at $L/d_p < 10$; however, for slightly oscillating drops it was not affected by vertical interaction over the range of $1.5 < L/d_p < 20$. By conducting experiments with single and triple nozzle arrangements, horizontal interaction effects were found to be negligible for both systems.

On a étudié les effets de l'interaction de gouttelettes circulantes ou oscillantes sur la formation de gouttes, la vitesse limite et le coefficient de transfert de masse en phase continue en chute libre. Pour examiner le coefficient de transfert de masse en une chute libre, on a éliminé les deux effets de bouts en soustrayant la masse transférée dans une colonne courte de celle transférée dans une colonne longue. On a mis au point une corrélation pour prédire le volume des gouttes, basée sur les équations de Scheele et Meister (1966) pour les gouttes non-éjectées, et sur les équations de Meister et Scheele (1969a, 1969b) pour gouttes formées dans les jets. La vitesse terminale des gouttes est augmentée par une interaction verticale avec les gouttes adjacentes pour des valeurs de $L/d_p < 5$. Le coefficient de transfert de masse en phase continue pour les gouttes circulantes est diminué par l'interaction verticale, lorsque $L/d_p < 10$; toutefois, dans le cas des gouttes légèrement oscillantes, ce coefficient n'est pas affecté par l'interaction verticale dans la gamme de $1{,}5 < L/d_p < 20$. En faisant des expériences avec des arrangements de gicleurs simples et triples, on a constaté que les effets de l'interaction horizontale étaient négligeables pour les deux systèmes.

Spray and perforated-plate columns are commonly used in commercial liquid–liquid extraction systems. In these towers many drops are formed simultaneously and rise or fall in a swarm. However, most studies have been done on systems employing single drops or a single stream of drops. Not much work has been undertaken to study the effects of neighboring drops on drop formation, terminal velocity and the mass transfer coefficient.

Desai (1980) studied the effects of interaction between oscillating droplets on formation, free fall, and mass transfer. This work was originally designed to further Desai's experiments by studying the same interaction effects for stagnant and circulating drops. During experimentation the system chosen to give stagnant drops was in fact found to yield drops exhibiting incipient oscillation. Besides these droplets, drops which circulated internally were studied. The drops were formed from nozzles immersed in an immiscible liquid medium. Drop size was investigated, as well as the terminal velocity of the free-falling drops, and the experimental values were compared with those predicted by existing correlations. The mass transfer coefficient in the continuous phase was measured and its variation due to neighboring drops investigated.

The work should aid in applying the design procedure for perforated plate extraction columns presented by Skelland and Conger (1973) to commercial systems, where dispersed phase holdups are quite high.

Relationships relevant to this study

Drop volume

The size of a drop formed in an immiscible liquid is dependent upon the physical properties of the system and the formation conditions. Scheele and Meister's (1968) correlation is the most accurate simple equation to use for drops formed under non-jetting conditions in systems of low viscosity liquids. It is written as:

$$V_F = F[(\pi\sigma d_n/g\Delta\rho) + (20\ \mu_c Q d_n/d_p^2 g\Delta\rho) - (4\ \rho_d Q U_n/3\ g\Delta\rho) + 4.5\ (Q^2 d_n^2 \rho_d \sigma/g^2\Delta\rho^2)^{1/3}] \quad \ldots\ldots (1)$$

The drag term (containing μ_c) may be neglected for continuous phase viscosities less than 0.01 Ns/m^2. This equation was derived for a parabolic velocity profile in the nozzle. For a flat velocity profile, one simply replaces the constant 4/3 in the kinetic term by 1.0.

In a later study Meister and Scheele (1969b) developed a correlation for drops formed under jetting conditions. Basing their analysis on a force balance they arrived at the following equations:

For jet lengths greater than one wavelength,

$$V_F = 2\pi^2 (a^3)_{\lambda/2}/N(Ka)_{max}(U_l/U_A)_{\lambda/2} \quad \ldots\ldots (2)$$

and for jet lengths less than one wavelength,

[1]Present address: Amoco Oil Company, 2401 5th Avenue South, Texas City, Texas 77590, USA.

$$V_F = F[(\pi\sigma d_j/g\Delta\rho) + (20\ \mu_c Q d_j/d_p^2 g\Delta\rho) - (4\rho_d Q U_n d_n^2/3 d_j^2 g\Delta\rho) + 7.15(Q^2 d_j^2 \rho_d \sigma/4 g^2 \Delta\rho^2)^{1/3}] \quad \ldots\ldots (3)$$

The above equation is very similar in form to Equation (1). In order to calculate d_j, the jet diameter, the length of the jet must be known. If this is not available it can be calculated by the equations of Meister and Scheele (1969a). The jet diameter can be predicted from Shiffler's jet contraction equation (Meister and Scheele, 1969a; Shiffler, 1965). This is an iterative procedure, but a reasonable estimate of d_p is usually sufficient for a quick solution.

Jetting velocity

As flow through a nozzle increases, a velocity U_{nj} will be reached at which jetting begins. Scheele and Meister (1968) proposed two mechanisms for jet formation. The first considers that a jet will be formed if there is a sufficient upward force at the nozzle exit. By making a balance on the interfacial tension, kinetic, and pressure forces, they obtained for a parabolic velocity profile in the nozzle,

$$U_{nj} = 1.73\ [\sigma(d_p - d_n)/\rho_d d_n d_p]^{0.5} \quad \ldots\ldots (4)$$

For a flat velocity profile in the nozzle the jetting velocity becomes

$$U_{nj} = 2.0\ [\sigma(d_p - d_n)/\rho_d d_n d_p]^{0.5} \quad \ldots\ldots (5)$$

The second mechanism considers that a jet will form when the velocity of the drop is low enough that it will rise less than one diameter during the time of formation of the next drop. The merging drops then form a continuous jet. The jetting velocity in this case is

$$U_{nj} = 2d_p^2 U_{Ri}/3d_n^2 \quad \ldots\ldots (6)$$

where U_{Ri} is the average rise velocity for the first diameter of rise.

Terminal velocity

When a drop is released from rest into another immiscible liquid it will accelerate to its terminal velocity and rise or fall through the liquid at this rate. Klee and Treybal (1956) noted a peak in the U_t versus d_p curve, corresponding to the beginning of drop oscillation. Their data were correlated by the following equations:

$$U_t = 3.042\rho_c^{-0.45}\Delta\rho^{0.58}\mu_c^{-0.11}d_p^{0.70}, \quad (d_p < d_c) \quad \ldots (7)$$

and

$$U_t = 4.96\rho_c^{-0.55}\Delta\rho^{0.28}\mu_c^{0.10}\sigma^{0.18}, \quad (d_p > d_c) \quad \ldots\ldots (8)$$

where

$$d_c = 1.772\rho_c^{-0.14}\Delta\rho^{-0.43}\mu_c^{0.30}\sigma^{0.24} \quad \ldots\ldots (9)$$

These expressions predict terminal velocities that increase with increasing drop diameter up to a critical diameter, after which the terminal velocity becomes constant, and is dependent only upon the physical properties of the system involved.

Grace et al. (1976) utilized data from several sources and conducted many experiments of their own, enabling their correlation to cover a wide range of variables. They found that the purity of a system had a large effect on the terminal velocity of a drop. Because of this they separated the data in the literature in accordance with the degree of care taken by the respective authors to eliminate surfactants. They developed the following correlation for contaminated systems:

$$J = 0.94H^{0.757} \quad (2 < H \leq 59.3) \quad \ldots\ldots (10)$$

$$J = 3.42H^{0.441} \quad (H > 59.3) \quad \ldots\ldots (11)$$

where

$$H = (4/3)N_{Eo}M^{-0.149}(\mu_c/\mu_w)^{-0.14} \quad \ldots\ldots (12)$$

$$J = N_{Re}M^{0.149} + 0.857 \quad \ldots\ldots (13)$$

The value of 59.3 for H corresponds to the transition from non-oscillating to oscillating drops. The terminal velocity is then given by

$$U_t = (\mu_c/\rho_c d_p)M^{-0.149}(J - 0.857) \quad \ldots\ldots (14)$$

For pure systems involving bubbles or drops at low Reynolds numbers, the above equation was modified to

$$U_{t,pure} = U_t[1 + 1/(2 + 3\mu_d/\mu_c)] \quad \ldots\ldots (15)$$

Continuous phase mass transfer coefficient

There are three stages during which mass transfer takes place to or from a single drop in an extraction column:

1) the formation stage;
2) the free fall period;
3) the coalescence period.

This work was concerned with the mass transferred during the free fall stage. When a drop falls through an immiscible liquid it will be either stagnant, internally circulating, or oscillating, and the mass transfer coefficient is thus dependent upon the state of the falling drops. Correlations for drops in each of these states are listed in Table 1.

For *swarms* of internally circulating drops rising or falling in a system of low interfacial tension, Treybal (1963) proposed the following correlation:

$$k_{cr} = 0.725(d_p U_s \rho_c/\mu_c)^{-0.43}(\mu_c/\rho_c D_c)^{0.58} U_s(1 - \phi_d) \quad \ldots\ldots (25)$$

where

$$U_s = U_d/\phi_d + U_c/(1 - \phi_d) \quad \ldots\ldots (26)$$

Criteria for drop oscillation

Some of the published criteria for the onset of droplet oscillation are:

a) $N_{We} > 3.58$	Hu and Kintner (1955)
b) $d_p > 1.772\ \mu_c^{0.30}\sigma^{0.24}/\rho_c^{0.14}\Delta\rho^{0.43}$	Klee and Treybal (1956)
c) $N_{Re}/N_{PG}^{0.15} > 20$	Johnson and Braida (1957)
d) $d_p > 0.162\ (\rho_d/\Delta\rho)^{0.5}$	Edge and Grant (1971)
e) $H > 59.3$	Grace et al. (1976)

Experimental work

Systems

The following continuous-phase-controlled systems were used in this work. The dispersed phase is specified first, followed by the solute and the continuous phase.

TABLE 1
Correlations for k_{cr} for Single Streams of Drops[a]

State of drop	Eqn. no.	Correlation	Reference
Stagnant[b]	16	$k_{cr} = 0.74(D_c/d_p)(N_{Re})^{1/2}(N_{Sc})^{1/3}$	Skelland and Cornish (1963)
Circulating[c]	17	$k_{cr} = (2/\sqrt{\pi})(D_c U_t/d_p)^{1/2}(8\phi'_{os\pi}/d_p^3)^{1/2}$	Shirotsuka and Kawase (1975, 1976)
Circulating[d]	18	$k_{cr} = (D_c/d_p)[2 + 1.13(N_{Re}N_{Sc})^{1/2}k_v^{1/2}]$	Griffiths (1960)
Circulating	19	$k_{cr} = (2/\sqrt{\pi})(D_c/d_p)\{1 - N_{Re}^{1/2} \times [2.89 + 2.15(\mu_d/\mu_c)^{0.64}]\}^{1/2}(N_{Re}N_{Sc})^{1/2}$	Weber (1975)
Circulating	20	$k_{cr} = (D_c/d_p)(-178 + 3.62N_{Re}^{1/2}N_{Sc}^{1/3})$	Thorsen and Terjesen (1962)
Circulating	21	$k_{cr} = (D_c/d_p)(-126 + 1.8N_{Re}^{1/2}N_{Sc}^{0.42})$	Garner et al. (1959)
Circulating	22	$k_{cr} = 0.6(D_c/d_p)N_{Re}^{1/2}N_{Sc}^{1/2}$	Garner and Tayeban (1960)
Oscillating	23	$k_{cr} = 1.2D_c^{1/2}[48\sigma/\pi^2 d_p^3(2\rho_c + 3\rho_d)]^{1/4}$	Clift et al. (1978), p. 197
Oscillating	24	$k_{cr} = (D_c/d_p)(50 + 0.0085N_{Re}^{1.0}N_{Sc}^{0.7})$	Garner and Tayeban (1960)

[a] N_{Re} and N_{Sc} refer to the continuous phase properties throughout.
[b] For other expressions, see Skelland (1974), pp. 276–277.
[c] $8\,\phi'_{os\pi}/d_p^3$ is given graphically as $f(L/D_p)$ in Shirotsuka and Kawase (1975).
[d] Selected from Griffiths' (1960) Table 2 since $(1 - k_v)(\xi/\xi_o)(k_v N_{Sc}^{1/3})^{-1} < 1/3$.

TABLE 2
Equilibrium Distribution

System	Relation	C^*_{Ac} range kmol/m^3
C_6H_5Cl + $C_6H_5CH_3$ $-C_6H_5COOH - H_2O$	$C^*_{Ad} = 7.868 \times 10^5 C^{*\,1.9904}_{Ac}$	$1.5 \times 10^{-3} - 1 \times 10^{-2}$
$H_2O - CH_3COOH - C_6H_5CH_3$	$C^*_{Ad} = 5.5463 \times 10^2 C^{*\,1.1377}_{Ac}$	$2 \times 10^{-4} - 1 \times 10^{-2}$

TABLE 3
Physical and Transport Properties

System	σ N/m	ρ_d kg/m^3	ρ_c kg/m^3	μ_d Ns/m^2	μ_c Ns/m^2	D_d m^2/s	D_c m^2/s	Temp. °C
C_6H_5Cl $C_6H_5CH_3$ $C_6H_5COOH -$ H_2O $H_2O - CH_3$	0.02746	1043.8	997.1	0.0006847	0.0008937[a]	not needed	1.2×10^{-9}[b]	25
$COOH - C_6H_5CH_3$	0.02225	997.1	866.0	0.0008937[a]	0.0006389	1.21×10^{-9}[c]	2.26×10^{-9}[c]	25

[a] From Skelland and Huang (1979).
[b] From Perry et al. (1963).
[c] From Chang and Wilke (1955).

System I: Mixture of Chlorobenzene (76.3 vol. %) and Toluene–Benzoic Acid–Distilled Water,

System II: Distilled Water–Acetic Acid–Toluene. The solute was transferred from the organic to the aqueous phase throughout. The distribution of the solute and the physical properties are shown in Tables 2 and 3. Chlorobenzene was supplied as "purified grade" with a residue after evaporation of 0.02%. The toluene used in system I was "Certified Reagent Grade" with a residue after evaporation of 0.0002%; for system II it was "Purified Grade". A "Fisher Certified Primary Standard" of benzoic acid was employed as solute in system I. Acetic acid, "Fisher Certified Reagent Grade", with a residue after evaporation of 0.0008% was used in system II. Volumetric analysis of solute was performed with sodium hydroxide solutions standardized daily against a solution of potassium biphthalate. To retain a single phase throughout a titration, 80 mL of isopropanol were added.

APPARATUS

The apparatus was designed to produce three parallel streams of drops. The general layout is described by Skelland and Caenepeel (1972) and by Skelland and Minhas (1971), and Figure 1 shows a diagram of the equipment. The principal modification in the present work compared to the two earlier papers consisted of the introduction of three Teflon needle valves (V_2, V_3, V_4), one for each nozzle, so that after each one had been adjusted to give a desired flow rate, the total flow could be turned on or off by valve V_1, without further adjustment of V_2, V_3 and V_4. The disperse phase was contained in a constant-head buret made from a 0.066 m ID glass column, with a height of 0.62 m.

The three nozzles were located on the corners of a 0.019 m equilateral triangle, and represented an attempt to simulate the hole distribution and multidrop formation conditions on a perforated plate. Six different nozzle sets were

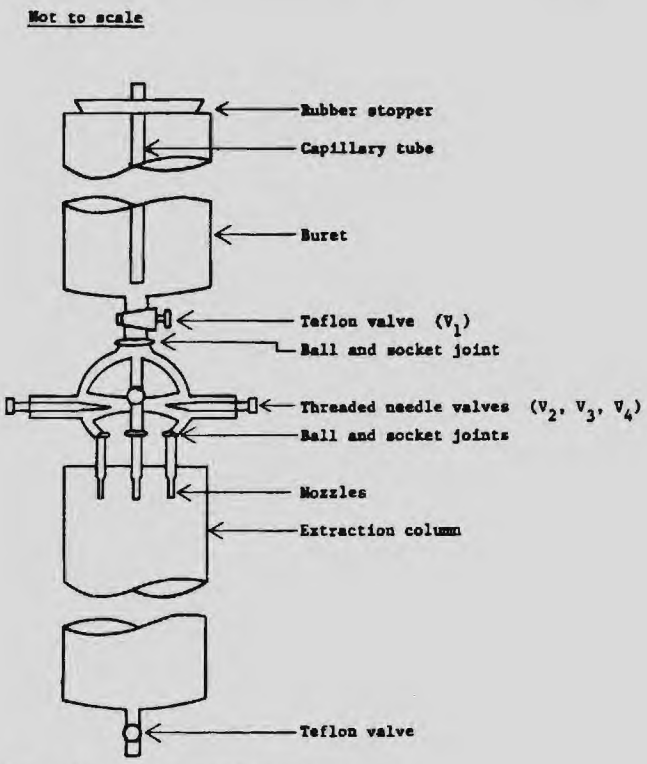

Figure 1 — Diagram of the apparatus.

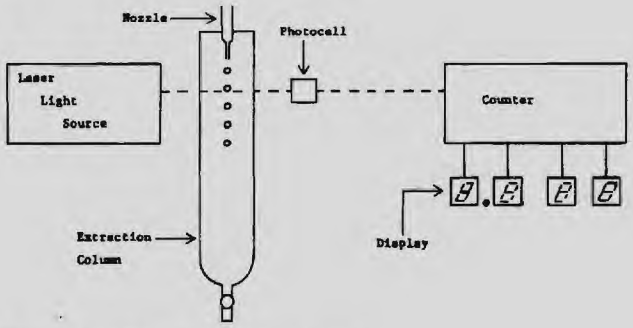

Figure 2 — Diagram of laser counter.

used, as described in Table 4. The nozzle tips were immersed in the stationary continuous phase, contained in a vertical cylindrical glass column of 0.047 m ID. The two column heights were 0.16 m and 0.857 m, respectively.

Measurements

Drop sizes were determined by counting the number of drops formed from a known volume of dispersed phase used during a given run. Depending on the nozzle rate, the formation times were measured either with a stopwatch or a photocell-digital counter. For drop formation times less than 0.40 s the digital counter assembly was used. The apparatus is illustrated in Figure 2. Briefly, a laser (L) was used to emit a beam of intense light which was detected by the photocell (P). Connected to the photocell was a digital counter (C) which gave the drop formation time in milliseconds. When a drop passed between the laser and the photocell it deflected the beam and this incident was recorded by the counter. The assembly counted ten drops and gave the average time per drop for the series of drops. The

TABLE 4
Dimensions of Nozzles

Nozzle set number	ID cm	OD cm	Length cm
1	0.0635	0.1631	3.9
2	0.0746	0.1598	4.0
3	0.0850	0.1962	4.4
4	0.1036	0.1884	3.8
5	0.0601	0.1507	3.9
6	0.1157	0.1951	3.8

terminal velocity of a drop was calculated by measuring the time that it took for a drop to fall 0.65 m. This was preceded by about 0.14 m of fall to allow the drop to reach its terminal velocity. Enough solute was dissolved to make the concentration in the dispersed phase (system I), or the continuous phase (system II), about 1% by weight. Exact concentrations of solute before and after each run were measured by titrating aliquot samples with an aqueous NaOH solution to a phenolphthalein end point. The overall mass transfer coefficient during free fall was then calculated as in the work of Garner and Skelland (1956), and Skelland and Wellek (1964).

By adjusting the drop formation time, the vertical distance between consecutive drops from each of the three nozzles could be varied. The range covered in this study was $1.20 \leq L/d_p \leq 45.2$ where d_p signifies the diameter of a sphere with the same volume as the drop and L is the vertical distance between consecutive drops. More detailed information concerning the operational, analytical, and computational procedures is given by Vasti (1981).

Results

State of the drops

The two classes of drops studied in this work were circulating and incipiently oscillating. In order to observe visually the state of the drops, tests were run with aluminum particles in the drops, which were allowed to fall through the appropriate continuous phase. Results of these tests indicated that system I yielded circulating drops. In similar trials the system II drops appeared to be stagnant for the low d_p region. Experimental results, however, indicated that these drops were actually in the range of incipient oscillation. Comparisons with the published criteria for the prediction of the onset of droplet oscillation are listed in Table 5. The drops were observed to fall through the column in a spiral path or in a zig-zag motion, as noted by previous authors (Clift et al., 1978, p. 188).

Drop volume

The drop volume for each system and nozzle size was plotted against the velocity of flow in the nozzle; two sample plots from the eight obtained appear in Figures 3 and 4. The predicted and observed jetting velocities are also indicated on these plots. These values are listed in Table 6. The drop volumes as predicted by Scheele and Meister's (1968) correlation (Equation (1)) for nozzle velocities below jetting are shown. For nozzle velocities in the jetting region, drop volumes as predicted by Meister and Scheele's (1969b) cor-

TABLE 5
Comparison Between Published Criteria for the Onset of Droplet Oscillation and the Ranges Encountered in this Work

Reference	System I		System II	
	Criterion	Range studied	Criterion	Range studied
Hu and Kintner (1955)	$N_{We} > 3.58$	$0.19 < N_{We} < 2.18$	$N_{We} > 3.58$	$0.83 < N_{We} < 3.92$
Klee and Treybal (1956)	$d_p > d_c$	$0.212 < d_p < 0.691$	$d_p > d_c$	$0.203 < d_p < 0.550$
Equation (9)	$d_c = 0.654\ \text{m} \times 10^2$		$d_c = 0.369\ \text{m} \times 10^2$	
Johnson and Braida (1957)	$N_{Re}/N_{PG}^{0.15} > 20.0$	$2.82 < N_{Re}/N_{PG}^{0.15} < 16.99$	$N_{Re}/N_{PG}^{0.15} > 20.0$	$7.30 < N_{Re}/N_{PG}^{0.15} < 26.12$
Edge and Grant (1971)	$d_p > d_{p,t}$ $d_{p,t} = 0.766\ \text{m} \times 10^2$	$0.212 < d_p < 0.691$	$d_p > d_{p,t}$ $d_{p,t} = 0.447\ \text{m} \times 10^2$	$0.203 < d_p < 0.550$

NOTE: Conclusion: System I is largely free from oscillation, but its drops were circulating. Many drops of System II were oscillating; incipient oscillation seems indicated.

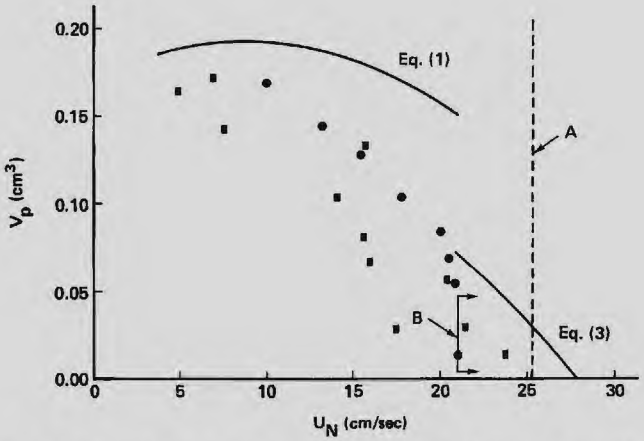

Figure 3 — Drop volume versus nozzle velocity. System I. Nozzle set 4 (●, single nozzle; ■, triple nozzle; A = predicted jetting, B = observed jetting).

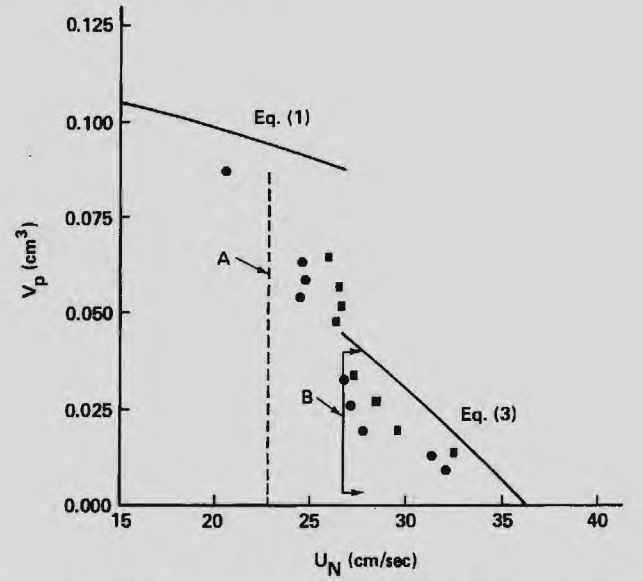

Figure 4 — Drop volume versus nozzle velocity. System II. Nozzle set 4 (●, single nozzle; ■, triple nozzle; A = predicted jetting, B = observed jetting).

relation (Equation (3)) are indicated. These values were calculated using the jet diameter at breakup as predicted by Skelland and Huang (1979), for d_j. The results indicate that there is an abrupt decrease in the drop volume at jetting.

It may be noted that drops of system I were observed to form at the nozzle internal diameter. In contrast, drops of system II formed at the outside diameter in the non-jetting region. These ID or OD values were used as appropriate when applying Scheele and Meister's (1968) drop volume correlation.

TABLE 6
Jetting Velocity

System	Nozzle ID cm	Nozzle velocity at jetting, cm/s	
		Predicted	Observed
System I	0.0635	32.97	29.23
System I	0.0746	30.23	24.82
System I	0.0850	28.15	23.99
System I	0.1036	25.26	21.01
System II	0.0601	30.91	40.52
System II	0.0850	25.75	32.99
System II	0.1036	22.94	26.86
System II	0.1157	21.50	24.52

It was desired to predict the drop volume by making a correction to the Scheele and Meister (1968) correlation to accommodate higher nozzle velocities. The drop volume data obtained by Desai (1980) have been incorporated in this portion of the research. Desai's two systems were as follows; the disperse phase is given first, followed by the solute and then the continuous phase.

System I. Chlorobenzene−Benzoic Acid−Water.
System II. Carbon Tetrachloride−Benzoic Acid−Water.
Initially the following relationship was proposed:

$$V_e/V_{sm} = f(N_{We,N}, N_{PG}) \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (27)$$

It was then assumed that

$$V_e/V_{sm} = K N_{We,N}^{\gamma} N_{PG}^{\beta} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (28)$$

The exponent γ was evaluated by plotting log (V_e/V_{sm}) versus log $(N_{We,N})$ for each system and obtaining the slope by linear regression analysis. These plots indicated different exponents depending on the region of flow (Vasti, 1981). Three regions were observed:

(1) Low $N_{We,N}$ region corresponding to low nozzle velocities.
(2) Intermediate $N_{We,N}$ region corresponding to a transition regime.
(3) High $N_{We,N}$ region corresponding to jetting velocities.

The region of each flow regime can be classified by the value of the $N_{We,N}$, as listed in Table 7, which also shows the values of γ for each flow region.

The next step was to evaluate β. This was accomplished by plotting log $(V_e/V_{sm}N_{We,N}^{\gamma})$ versus log (N_{PG}), using the appropriate value of γ, and calculating the slope through

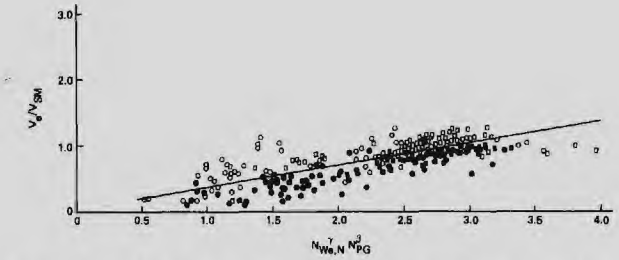

Figure 5 — Multiple (triple) nozzle drop volume correlation: V_e/V_{sm} versus $N_{We,N}{}^{\gamma}N_{PG}{}^{\beta}$ (●, System I; ○, System II; □, System I Desai (1980); ■, System II Desai (1980)).

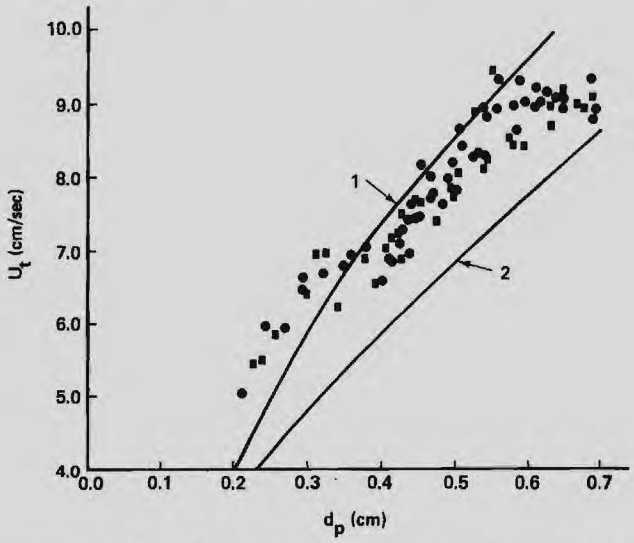

Figure 6 — Terminal velocity versus drop diameter. System I (●, single nozzle; ■, triple nozzle; 1 = correlation of Grace et al. (1976); 2 = correlation of Klee and Treybal (1956)).

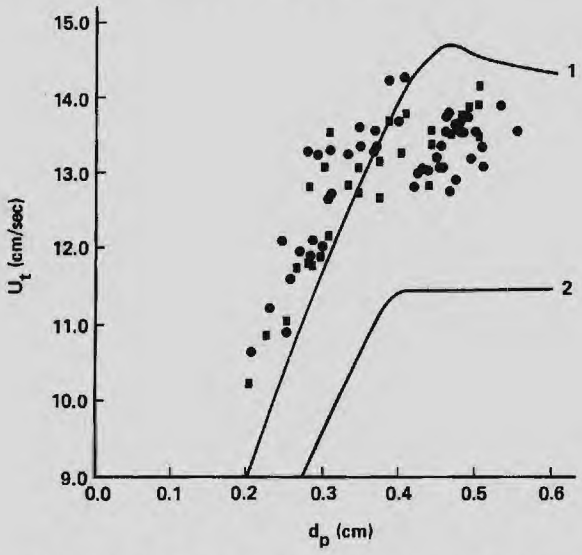

Figure 7 — Terminal velocity versus drop diameter. System II (●, single nozzle; ■, triple nozzle; 1 = correlation of Grace et al. (1976); 2 = correlation of Klee and Treybal (1956)).

these points. Two values of β were obtained; one for the non-jetting region, and one for the jetting region. These values and regions of flow are listed in Table 7.

The final step was to evaluate the constant, K. This was done by plotting log (V_e/V_{ms}) versus log $(N_{We,N}{}^{\gamma}N_{PG}{}^{\beta})$, using the correct values of γ and β, and calculating the slope

TABLE 7
Values for Drop Volume Correlation

$N_{We,N}$ region	γ	β
$0 < N_{We,N} < 0.7$	−0.04758	0.03691
$0.7 < N_{We,N} < 1.75$	−0.75070	0.03691
$N_{We,N} > 1.75$	−2.23034	0.14540

through these points. This is plotted in Figure 13. Linear regression was used to obtain a value of 0.3078 for K. The final correlation can be expressed as

$$V_e/V_{sm} = 0.3078\, N_{We,N}{}^{\gamma}N_{PG}{}^{\beta}, \quad 0.5 \le N_{We,N}{}^{\gamma}N_{PG}{}^{\beta} \le 4.0 \qquad (29)$$

where γ and β are given in Table 7.

TERMINAL VELOCITY

The terminal velocities for both systems are plotted versus drop diameter d_p in Figures 6 and 7. Also shown are the terminal velocities for these drops falling singly as predicted by the correlations of Klee and Treybal (1956) (Equations (7−9)) and Grace et al. (1976) (Equations (10−15)). The latter correlation, which is for very pure systems, fits the data of both systems better than that of Klee and Treybal (1956). In both plots the data follow the shape of the Klee and Treybal correlation, but are all higher. In their work, Klee and Treybal (1956) did not take great care to eliminate impurities in their materials, yet the presence of surfactants has since been shown to reduce droplet terminal velocities (Skelland and Huang, 1977). The presence of parallel droplet streams appears to have no noticeable effect on the terminal velocity of the drops. During these experiments the drops seldom fell side by side so that this effect would be difficult to study. The terminal velocity is observed to increase with increasing droplet diameter, while in system II the terminal velocity reaches a maximum. This is due to the onset of oscillations and has been observed by previous authors (Johnson and Braida, 1957; Klee and Treybal, 1956). The scatter of the data can be attributed to the somewhat erratic drifting of the drops and the zig-zag or spiral paths of the drops as previously described.

In order to study any possible interaction effect, it was necessary to eliminate the effect of droplet diameter on terminal velocity. This was accomplished in the following manner. For a given system, from Klee and Treybal's (1956) correlation for d_p less than d_c,

$$U_{t,d_p} \propto d_p{}^{0.70} \qquad (30)$$

If $d_p \ge d_c$ then U_{t,d_p} is a constant value. Therefore a representative or "standard" drop diameter can be chosen. This was selected as the largest drop diameter observed in either system. It follows from Equation (30) that

$$U_{t,d_{std}} = U_{t,d_p}(d_{std}/d_p)^{0.70} \qquad (31)$$

By plotting $U_{t,d_p}(d_{std}/d_p)^{0.70}$ versus L/d_p as in Figures 8 and 9 possible interaction effects can be observed.

Figure 8 indicates that there is some interaction present at $L/d_p < 20$. The most significant interaction effects are observed at $L/d_p < 5$. As the vertical distance between drops increases for $L/d_p > 20$, the drops behave as single drops and no interaction is present.

For system II, most of the data are in the low L/d_p region, as evidenced by Figure 9, which contains only those cases

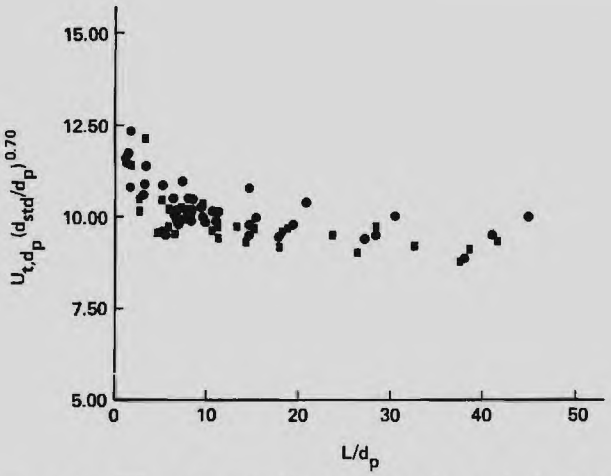

Figure 8 — Effects of vertical interaction on the terminal velocity for circulating drops: $U_{t,d_p}(d_{std}/d_p)^{0.70}$ versus L/d_p. System I (●, single nozzle; ■, triple nozzle).

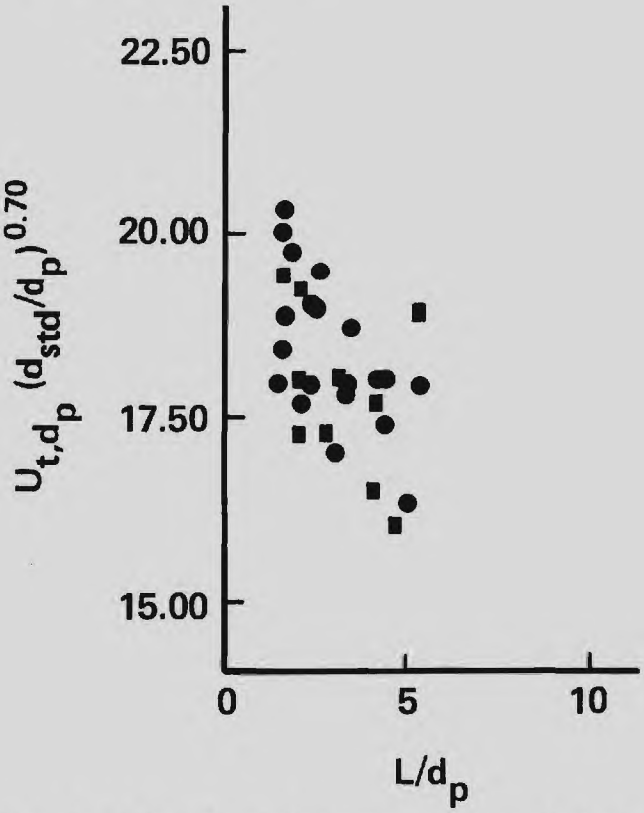

Figure 9 — Effects of vertical interaction on the terminal velocity for incipiently oscillating drops: $U_{t,d_p}(d_{std}/d_p)^{0.70}$ versus L/d_p. System II (●, single nozzle; ■, triple nozzle).

where $d_p < d_c$. For this system appreciable interaction effects are observed for $L/d_p < 5$. These plots indicate that the terminal velocity is enhanced by the presence of vertically neighboring drops at low L/d_p. It was often observed that a drop falling closely behind another would pass the leading drop, eventually slow down and sometimes be passed by the newly trailing drop. This phenomenon was sometimes observed over a length of 20−30 cm.

Figures 8 and 9 indicate enhancement in drop velocity of about 20% when L/d_p is reduced from 5 to 2. Although not generalized, this figure shows the order of magnitude that may be expected in terminal velocity effects due to vertical interaction. A correctional procedure for swarms of drops might therefore proceed in the following tentative manner:

1) Compute d_p from Equation (29).
2) Estimate u_t from the correlation of Grace et al. (1976) (Equations (10−15)) if the system is very pure, or from Klee and Treybal (1956) (Equations (7−9)) if "normal degrees" of contamination are anticipated.

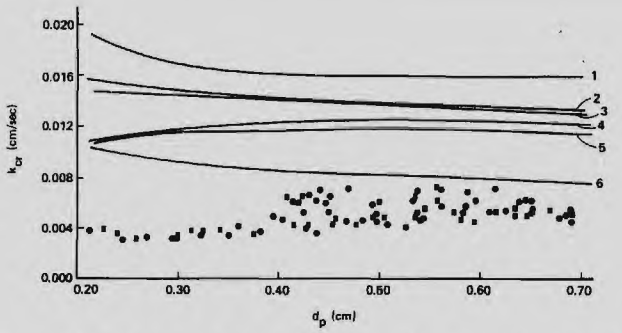

Figure 10 — k_{cr} versus drop diameter. System I (circulating drops) (●, single nozzle; ■, triple nozzle; curves 1 to 6 are the correlations of Shirotsuka and Kuwase (1976), Griffiths (1960), Weber (1975), Thorsen and Terjesen (1962), Garner et al. (1959), and Garner and Tayeban (1960), respectively).

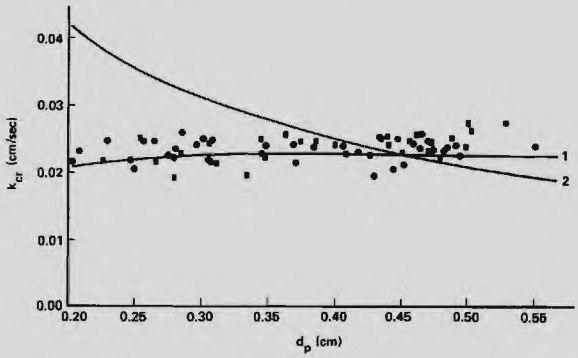

Figure 11 — k_{cr} versus drop diameter. System II (incipiently oscillating drops) (●, single nozzle; ■, triple nozzle; 1 = correlation of Garner and Tayeban (1960); 2 = correlation of Clift et al. (1978)).

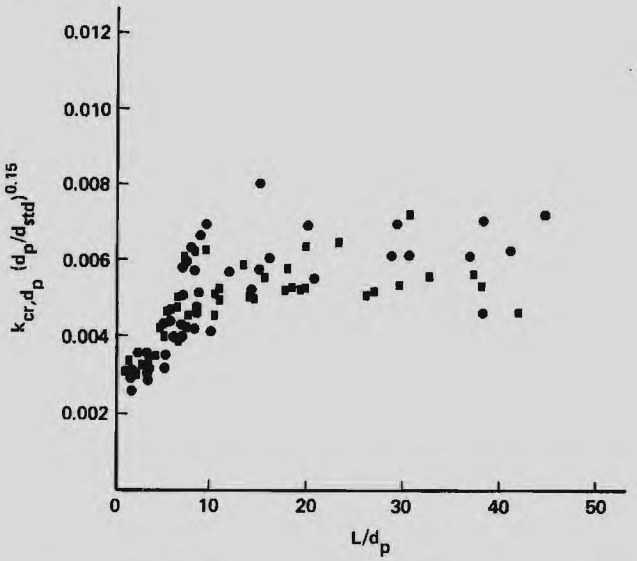

Figure 12 — Effects of vertical interaction on the continuous phase mass transfer coefficient for circulating drops: $k_{cr,d_p}(d_p/d_{std})^{0.15}$ versus L/d_p. System I (●, single nozzle; ■, triple nozzle).

3) Calculate the drop formation time as $t_f = 2d_p^3/3d_n^2u_n$.
4) Obtain a first estimate of L/d_p as $u_t t_f/d_p = (2/3)\cdot(u_t/u_n)(d_p/d_n)^2$.
5) If L/d_p in (4) is >5, retain the u_t estimate from (2) and the L/d_p from (4).
6) If L/d_p in (4) is <5, estimate the % increase in u_t due to vertical interaction from Figure 8 or 9. (This assumes, as an expedient, that the % change in u_t over a given range of L/d_p is independent of the system.)
7) Repeat steps (4−6) until u_t and L/d_p become constant.

MASS TRANSFER DURING FREE FALL

The continuous phase mass transfer coefficients for both systems are plotted versus drop diamter d_p in Figures 10 and 11. Also shown in Figure 10 are the values predicted by the correlation of Shirotsuka and Kawase (1976), which allows for vertical interaction, but indicates that this is significant only for $L/d_p \leq 4$. In addition, predictions are indicated from the circulating drop relationships presented by Griffiths (1960, his Table 2, formula 4), Weber (1975), Thorsen and Terjesen (1962), Garner et al. (1959), and Garner and Tayeban (1960), none of which accounts for droplet interaction. All of these expressions are listed in Table 1. Figure 10 shows that k_{cr} decreases slightly with decreasing d_p. This is at variance with most of the relationships plotted in Figure 10, including that of Garner and Tayeban, to which the data are closest, suggesting some vertical interaction effect which reduces the continuous phase coefficient (L/d_p was smaller with smaller d_p values). No interaction between parallel drop streams is apparent, since no stratification is seen between the single and triple nozzle data points.

Figure 11 shows that the incipiently oscillating drop data (system II) are well fitted throughout by the correlation of Garner and Tayeban (1960) for oscillating drops, while showing regions of deviation from the expression given for oscillating drops by Clift et al. (1978). The Garner and Tayeban (1960) correlation shown in Figure 11 is based on data from three experimental systems and contains no allowance for vertical interaction. The agreement exhibited indicates the absence of such interaction in the incipient oscillation range. The absence of interaction due to parallel drop streams again follows from the lack of stratification between the single and triple nozzle data.

As in the terminal velocity studies, it was necessary to eliminate the effect of droplet diameter on k_{cr} for circulating drops as follows. For a given system, Equation (22) with Equation (30) shows that, for circulating drops,

$$k_{cr,d_p} \propto d_p^{-0.15} \quad \ldots\ldots (32)$$

As before, a "standard" drop diameter was chosen as the largest drop diameter observed. It follows from Equation (32) that

$$k_{cr,d_{std}} = k_{cr,d_p}(d_p/d_{std})^{0.15} \quad \ldots\ldots (33)$$

By plotting $k_{cr,d_p}(d_p/d_{std})^{0.15}$ versus L/d_p as in Figure 12 significant interaction effects are seen for $L/d_p < 10$. This shows that for circulating drops in the range of $L/d_p < 10$ the mass transfer coefficient is reduced due to the presence of neighboring drops in a vertical stream.

Figure 12 indicates a reduction in the continuous phase mass transfer coefficient of about 45% when L/d_p is reduced from 10 to 2. Although not generalized, this may give some rough guidance on the effects of vertical interaction on k_{cr}. Thus a tentative correction procedure for swarms of drops might parallel that already described for terminal velocity. The u_t and L/d_p estimated there in a 7-step computation may be used in the correlation chosen to calculate k_{cr} for circulating drops, and the % reduction in this k_{cr} due to vertical interaction is then estimated using Figure 12. Again the expedient assumption is made that the % change in k_{cr} over a given range of L/d_p is independent of the system.

Conclusions

The following conclusions can be drawn from this study:
1) The drop volume has been correlated using modifications of the correlations of Scheele and Meister (1968) for non-jetting drops, and Meister and Scheele (1969b) for drops formed under jetting conditions.
2) The correlation of Grace et al. (1976) was found to best predict terminal velocities.
3) The continuous phase mass transfer coefficient for circulating drops was best predicted by the appropriate correlation of Garner and Tayeban (1960).
4) For oscillating drops, the continuous phase mass transfer coefficient was best predicted by the appropriate correlation of Garner and Tayeban (1960).
5) The terminal velocities for both systems were enhanced by vertical interaction with adjacent drops for values of $L/d_p < 5$. No horizontal interaction was observed.
6) The continuous phase mass transfer coefficient for circulating drops was reduced due to vertical interaction at $L/d_p < 10$. No effect due to horizontal interaction was detected.
7) For incipiently oscillating drops, no interaction effects on the continuous phase mass transfer coefficient were observed.

Acknowledgements

Some of the drop volume measurements used here were made by T. M. Desai at the University of Kentucky. The work was partially supported by National Science Foundation Grant Nos. ENG 77-14609, CPE 80-19617, and CPE-8203872.

Nomenclature

a = jet radius, m
C^*_{Ac}, C^*_{Ad} = equilibrium solute concentration in continuous and dispersed phases, respectively, kmol/m^3
D_c, D_d = molecular diffusivity of solute in continuous and dispersed phases, m^2/s
d_c = critical drop diameter defined in Equation (9), m
d_j = jet diameter, m
d_N = nozzle diameter (ID or OD as appropriate), m
d_p = diameter of sphere with the same volume as the drop, m
$d_{p,t}$ = transition drop diameter, m
d_{std} = standard drop diameter, m
Eo = Eotvos number = $g\Delta\rho d_p^2/\sigma$
F = Harkins-Brown factor
f_1 = function
g = acceleration due to gravity, 9.81 m/s^2
g_c = gravitational constant
H = dimensionless group defined by Equation (12)
J = dimensionless group defined by Equations (10, 11)
K = constant in Equation (28)
$(K_a)_{max}$ = dimensionless wave number of dominant symmetrical wave
k_{cr} = individual mass transfer coefficient for continuous phase during free rise or fall, m/s

k_{cr,d_p} = individual mass transfer coefficient for continuous phase during free fall for a drop of diameter d_p, m/s

$k_{cr,d_{std}}$ = individual mass transfer coefficient for continuous phase during free fall for a drop of "standard" diameter, m/s

k_v = ratio of actual interfacial speed to the potential flow interfacial speed

L = vertical distance between two consecutive drops, m

M = modified Morton number = $g\mu_c^4\Delta\rho/\rho_c^2\sigma^3$

N = factor in Equation (2) to account for wave splitting

N_{PG} = physical property group = $(\rho_c^2\sigma^3 g_c^3)/(g\mu_c^4\Delta\rho)$

N_{Re} = Reynolds number = $U_t d_p \rho_c/\mu_c$

N_{We} = Weber number = $(d_p U_t^2 \rho_c)/(\sigma g_c)$

$N_{We,N}$ = nozzle Weber number = $(U_n^2 d_n \rho_d)/(\sigma g_c)$

Q = volumetric flow rate, m^3/s

U_A = average jet velocity, m/s

U_c, U_d = superficial velocity of continuous and dispersed phases based on empty column cross section, m/s

U_I = interfacial velocity, m/s

U_n = velocity of the dispersed phase through the nozzle, m/s

U_{nj} = jetting velocity through the nozzle, m/s

U_{Ri} = average rise velocity of drop over first drop diameter of rise, m/s

U_s = slip velocity, m/s

U_t = terminal velocity, m/s

U_{t,d_p} = terminal velocity of a drop of diameter d_p, m/s

$U_{t,d_{std}}$ = terminal velocity of a drop of "standard" diameter, m/s

$U_{t,pure}$ = terminal velocity of a drop or bubble in the limit of absolute purity, i.e., for no surface-active contaminant present, m/s

V_e = experimental value of drop volume, m^3

V_F = volume of forming drop, m^3

V_p = volume of drop, m^3

V_{sm} = volume of drop calculated by Equation (1) due to Scheele and Meister (1968), m^3

Greek letters

β = exponent in Equation (28)

γ = exponent in Equation (28)

Δ = difference

μ_c, μ_d = viscosity of the continuous and dispersed phases, respectively, Ns/m^2

μ_w = viscosity of water, Ns/m^2

ξ/ξ_0 = ratio of the concentration boundary layer thickness with interfacial flow to that without interfacial flow

ρ_c, ρ_d = density of the continuous and dispersed phases, respectively, kg/m^3

σ = interfacial tension, N/m

ϕ_d = dispersed phase hold up

$\phi_{os\pi}$ = approximate velocity potential at stagnation point, m^3

Subscripts

$\lambda/2$ = one half of wavelength, m, at a distance

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Minimum Agitator Speeds for Complete Liquid–Liquid Dispersion

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Data from three different sources have been pooled to obtain an empirical correlation of the minimum agitator speed needed to obtain complete liquid–liquid dispersion in baffled vessels. The variables include 5 common types of impellers, 2 generating axial flow and three radial flow, in four locations, fluid properties in 11 systems, tank diameter, liquid height, and volume fraction of the disperse phase.

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Observations from 481 runs were correlated with an average absolute deviation of 12.7% by the expression $(N_{Fr})_{min} = C^2(T/D)^{2\alpha}\phi^{0.105}(N_{Ga}N_{Bo})^{-0.084}$ where C and α are tabulated according to the impeller type, location, and—in some cases—the ratio H/T. Scale-up relationships are developed from the above expression.

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Liquid–liquid dispersion in agitated vessels finds extensive application in mixer–settler design in extraction operations and in emulsion polymerization processes. In such work, it is necessary to ensure that the agitator speed is high enough to achieve complete dispersion of one liquid in the other. Skelland and Seksaria (1978) showed that, in some cases, impeller speeds of 1000 rpm are insufficient to ensure complete dispersion.

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The few previous studies began with Nagata's (1950) work; he used an unbaffled, flat-bottomed vessel, with a centrally mounted, four-bladed flat-blade turbine agitator, with T/D of 3 and a blade width of $0.06T$. He obtained the empirical expression

$$N_{min} = 6D^{-2/3}\left(\frac{\mu_c}{\rho_c}\right)^{1/9}\left(\frac{\Delta\rho}{\rho_c}\right)^{0.26} \quad (1)$$

REQU 1 (006, 6– 7)

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and he reported that N_{min} is independent of interfacial tension.

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Van Heuven and Beek (1971) studied dispersion with a six-bladed disk turbine in a baffled vessel. From a combination of theory and experiment, they obtained

$$N_{min} = \frac{3.28g^{0.38}\Delta\rho^{0.38}\mu_c^{0.06}\sigma^{0.06}(1 + 2.5\phi)^{0.90}}{D^{0.77}\rho_M^{0.54}} \quad (2)$$

show "ϕ" in same type as in equations 6, 7, 8 ... 13.

REQU 2 (006,10–11)

11

where D/T was constant at 0.3.

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The study by Skelland and Seksaria (1978) included a variety of impellers–propellers, pitched-blade turbines, flat-blade turbines, and curved-blade turbines—in a baffled vessel. They found

$$N_{min} = C_0D^{\alpha_0}\mu_c^{1/9}\mu_d^{-1/9}\sigma^{0.3}\Delta\rho^{0.25} \quad (3)$$

REQU 3 (006, 3– 4)

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where $H/T = 1$ and $\phi = 0.5$; T did not vary. The constants C_0 and α_0 were functions of the impeller type and location.

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A related phenomenon is the minimum impeller speed required to suspend solid particles off the bottom of the vessel in an agitated solid–liquid suspension. Zwietering (1958) used turbines, paddles, propellers, and vaned disks to suspend sand and sodium chloride particles in liquids in baffled vessels. He presents

$$N_{min} = C'\left(\frac{T}{D}\right)^{\alpha'}\frac{g^{0.45}\Delta\rho^{0.45}\mu_c^{0.1}D_p^{0.2}(100R)^{0.13}}{D^{0.85}\rho_c^{0.55}} \quad (4)$$

$D_p^{0.2}$

REQU 4 (009, 3– 4)

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Pavlushenko et al. (1957) used three-bladed square-pitch propellers for sand and iron suspensions to obtain

$$N_{min} = 0.105\left(\frac{T}{D}\right)^{1.9}\frac{g^{0.8}\rho_d^{0.8}D_p^{0.4}}{D^{0.6}\rho_c^{0.6}\mu_c^{0.2}} \quad (5)$$

REQU 5 (003,16–17)

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Differences in the exponents on a given variable in eq 4 and 5 are noteworthy, including the conflicting directional effect of μ_c indicated by the two expressions.

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SEN03 1 **Experimental Apparatus and Procedure**

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SEN03 1 **Fluids Used.** Deionized water formed one phase in all
SEN06 9 runs; the other phase consisted of one of the following
19 presaturated with water in all cases: ethyl acetate, benz-
28 aldehyde, chlorobenzene, and carbon tetrachloride.

PAR30

SEN03 1 Fluid properties appear in Table I. Interfacial tensions
SEN06 4 for ethyl acetate and benzaldehyde are from Skelland and
13 Seksaria (1978), chlorobenzene from Moeti (1984), and
20 carbon tetrachloride from the International Critical Tables
27 (1928).

TBL I (003, 7– 8)

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SEN03 1 **Apparatus.** The Experimental Agitator Model ELB
SEN06 7 manufactured by Bench Scale Equipment Co. was used
SEN09 15 for the liquid–liquid dispersion studies. The unit included
5 a $^1/_4$-hp drive motor, which supplied an infinitely variable
SEN12 14 output speed of 0–20 rps. The accuracy of the speed
7 control dial was checked with a stroboscope.

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SEN03 1 Three glass, cylindrical, flat-bottomed vessels were used,
9 providing the ranges $2.13 \leq T/D \leq 3.83$ and $^1/_2 \leq H/T$
SEN06 21 $\leq {}^3/_2$. Four radial baffles at 90° intervals were employed,
SEN09 10 with $T/B = 12$. The continuous phase was identified with
8 the aid of two conductivity electrodes made of copper wire,
18 0.01 m apart and connected to a conductivity meter.

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SEN03 1 Agitation was supplied by three sizes of a centrally
11 mounted six-flat-bladed turbine on a concentric vertical
SEN06 18 shaft. The shaft, impellers, and baffles were all made of
SEN09 11 316 stainless steel. Dimensions of the equipment are listed
8 in Table II, and a diagram is shown in Figure 1.

TBL II (009,10–11), FIG 1 (009,18–19)

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SEN03 1 **Operational Procedure.** Before filling with a given
SEN06 7 pair of liquids, the equipment was washed with detergent
16 and then rinsed with tap water, followed by deionized
26 water.

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SEN03 1 Two immiscible liquids in the desired volume propor-
SEN06 9 tions ($0.1 \leq H_2O \leq 0.9$) were added to the vessel. The
3 impeller was centrally located at $H/2$ in all runs, and its
14 speed gradually increased in small increments, until visual
22 observation showed that complete dispersion was just
SEN09 29 achieved. This state has been defined by Skelland and
10 Seksaria (1978) as follows: "The minimum mixing speed
19 of the impeller is the rotational speed just sufficient to
29 completely disperse one liquid in the other, so that no clear
40 liquid is observed either at the top or the bottom of the
SEN12 52 mixing vessel. In some cases, clear liquid pockets of ap-
9 proximately 1×10^{-6} to 5×10^{-6} m^3 persisted near the sides
22 of the vessel at the top and the bottom, although the rest
SEN15 34 of the liquids were mixed. In some instances, small pools
7 of clear liquid adhered to the drive shaft at the top of the
SEN18 30 vessel. In order to mix these small liquid pockets, speeds
SEN21 11 had to be increased by 25–100% or more. To prevent such
5 anomalous results, a well-mixed or completely dispersed
12 state was defined when only small, relatively nonstationary,
20 liquid pockets remained unmixed in the bulk dispersion.
SEN24 1 The rotational speed of the impeller corresponding to this
11 state is defined as the minimum mixing speed, N_{min}, and
21 is not necessarily the same as that required for a ho-
SEN27 31 mogenous dispersion. More nearly homogenous disper-
5 sions may have occurred at speeds higher than the mini-
14 mum mixing speed, in accordance with the findings of
23 Pavlushenko (1957) and Zwietering (1958)".

PAR48

SEN03 1 In the range $0.4 \leq \phi \leq 0.6$, after the continuous phase
14 had been identified in a given run, phase inversion was
24 achieved in the next run by initiating dispersion with an
34 auxilary off-center turbine, temporarily placed in the phase
SEN06 42 desired to be continuous. The auxiliary turbine was
6 withdrawn after dispersion was established, the centrally
13 mounted turbine being started just prior to this with-
21 drawal, so as to maintain the dispersion.

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SEN03 1 The reproducibility of the minimum mixing speed
9 needed to achieve complete dispersion was good and was
18 quantitatively similar to that found by Skelland and
26 Seksaria (1978).

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SEN03 1 The identification of the continuous phase in a given run
12 was usually possible by the visual methods described in
21 detail by Quinn and Sigloh (1963) and by Selker and
SEN06 31 Sleicher (1965). However, the most reliable means of
8 identification was provided by the conductivity electrodes
16 in the vessel, as described earlier, and connected to a
SEN09 25 conductivity meter. The conductivity of the aqueous phase
8 was much greater than that of the organic liquids, making
SEN12 18 continuous phase identification a simple matter. Data and
4 results for each run are recorded by Ramsay (1984).

TXT12

SEN03 1 **Correlation of Results**

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SEN03 1 To increase the generality of the correlation to be de-
11 veloped, the results from the present 251 runs were com-
20 bined with those from 35 runs reported by van Heuven and
31 Beek (1971) and from the 195 runs made by Skelland and
SEN06 42 Seksaria (1978). This gave a total of 481 data points on
11 5 types of impeller (3 radial flow and 2 axial flow), 4 im-
23 peller locations, and 11 systems, with $0.01 \leq \phi \leq 0.6$,
SEN09 35 $\leq T/D \leq 3.83$, and $1/2 \leq H/T \leq 3/2$. The impellers fea-
4 tured in the combined work are shown in Figure 2.

FIG 2 (009,13–14)

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SEN03 1 The data were correlated by using the BMDP Statistical
11 Software, page 264, Program 9R, University of California
19 Press, 1983.

PAR63

SEN03 1 In accomodating the densities of the two phases, van
11 Heuven and Beek (1971) were successful in using a mean
21 density defined as

$$\rho_M = \phi\rho_d + (1 - \phi)\rho_c \qquad (6)$$

REQU 6 (003,23–24)

PAR66

SEN03 1 This same expression was used by Laity and Treybal
11 (1957) in correlating power consumption in agitated liq-
18 uid–liquid systems in baffled vessels, together with the
26 Vermeulen et al. (1955) expression for mean viscosity,
34 which is

$$\mu_M = \frac{\mu_c}{1 - \phi}\left(1 + \frac{1.5\mu_d\phi}{\mu_d + \mu_c}\right) \qquad (7)$$

REQU 7 (003,35–36)

SEN06 1 In the present work, N_{min} was the dependent variable,
11 whereas H, T, D, $\Delta\rho$, ρ_M, μ_M, σ, and ϕ were independent
23 variables; ρ_M and μ_M are given by eq 6 and 7, respectively.

PAR69

SEN03 1 The pooled 481 runs provided 16 useable sets of data,
12 corresponding to 5 different impellers in 4 different loca-
20 tions (including 2 impellers on a shaft) and with H/T
30 assuming values of $^1/_2$, 1, and $^3/_2$ for the centrally mounted
SEN06 41 flat-blade turbine. For all the other impellers H/T was
SEN09 9 unity. Our correlation is as follows, based on the dimen-
10 sional analysis, reformulation of the resulting groups in
18 terms of appropriate force ratios (Boucher and Alves,
26 1959), and using an assumed exponential form

$$\frac{N_{min}^2\rho_m^2}{g\Delta\rho} = C^2\left(\frac{T}{D}\right)^{2\alpha}\phi^{0.106}\left(\frac{\mu_m^2\sigma}{D^5\rho_m^2\Delta\rho^2}\right)^{0.084} \qquad (8)$$

REQU 8 (009,32–33)

SEN12 1 The correlation coefficient is 0.92 and the 95% confidence
11 intervals on the last two exponents are 0.106 ± 0.073 and
SEN15 22 0.084 ± 0.017. (The presence of ϕ in ρ_M and μ_M may
11 account for the small residual dependence on this variable
SEN18 20 shown in eq 8.) The experimental ranges covered for the
8 dimensionless groups were $1.75\ (10^7) \leq (N_{Gr}N_{Bo}) \leq 8.78$
SEN21 17 (10^{12}) and $0.17 \leq N_{Fr} \leq 38.8$. The values of C and α, with
9 the attendant 95% confidence intervals on α for the var-
SEN24 18 ious geometries studied, are given in Table III. This table
4 also shows the average percent deviation between calcu-
11 lated and experimental N_{min}, defined as $100|N_{min,calcd} -$
19 $N_{min,exptl}|/N_{min,calcd}$.

TBL III (021,25–26)

SEN03 1 The exponent on T/D for the flow number
12 to be the same as that determined by Zwietering (1978)
22 for solid–liquid dispersions; his impeller and vessel di-
29 mensions were similar to those of van Heuven and Beek
SEN06 39 (1971). This was done because the range of T/D used by
12 van Heuven and Beek was not sufficient to achieve a re-

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liable exponent (T/D equaled 3 for four systems and 3.33 for the other system).

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SEN03 1 The only check for this assumption is the exponent on
SEN06 12 T/D for a propeller located at $H/4$. Zwietering's exponent
SEN09 4 was 0.90 compared to the 0.67 of Seksaria. Because the
4 values are reasonably close, it seems safe to assume that
14 the exponent for T/D is the same for the dispersion of
25 liquids as for the suspension of solids, at least until further
26 experimental data are taken.

PAR78

SEN03 1 The last dimensionless group on the right-hand side of
11 eq 8 is the product of the reciprocals of the Galileo and
SEN06 23 the Bond numbers. The former (N_{Ga}) is proportional to
8 the ratio of inertial times gravitational force to the square
SEN09 18 of the viscous force in the system. It has previously found
6 application in studies on circulation in baths of viscous
SEN12 15 liquids (Boucher and Alves, 1959; Kruszewski, 1957). The
3 Bond number is proportional to the ratio of gravitational
SEN15 12 to interfacial tension forces prevailing in the system. It
3 has previously been used in work on the atomization of
13 liquids into droplets (Boucher and Alves, 1959; Richardson,
SEN18 21 1953). The group on the left-hand side of eq 8 is the
13 Froude number, proportional to the ratio of inertial to
SEN21 22 gravitational forces. These considerations enable eq 8 to
6 be expressed in the abbreviated form

$$\left(N_{Fr}\right)_{min} = C^2\left(\frac{T}{D}\right)^{2a}\phi^{0.108}(N_{Ga}N_{Bo})^{-0.084} \qquad (9)$$

REQU 9 (021,13–14)

PAR81

SEN03 1 Expansion of eq 8 yields the expression

$$N_{min} = C\left(\frac{T}{D}\right)^{a}\frac{g^{0.42}\Delta\rho^{0.42}\mu_m^{0.08}\sigma^{0.04}\phi^{0.06}}{D^{0.71}\rho_M^{0.54}} \qquad (10)$$

REQU 10 (003, 8– 9)

PAR84

SEN03 1 Comparison between eq 10 and Zwietering's relationship
9 for solid–liquid suspensions in eq 4 is consistent with the
19 statement by van Heuven and Beek (1971) to the effect
29 that "there are no good reasons to suppose that a great
40 difference exists between the mechanism of "complete
47 dispersion" (liquid–liquid) and "complete suspension"
52 (solid–liquid)".

PAR87

SEN03 1 Equation 10 also compares closely in many respects with
11 eq 2 for liquid–liquid dispersion using six-bladed disk
19 turbine impellers, as given by van Heuven and Beek (1971).
SEN06 1 In their work, T/D was constant at 3.33 and was therefore
13 a part of their coefficient 3.28.

PAR90

SEN03 1 Figures 3, 4, and 5 show plots of $N_{min,calcd}$ vs. $N_{min,expt}$
13 for the data sets provided by Skelland and Seksaria, van
23 Heuven and Beek, and the present work, respectively.

FIG 3 (003, 3– 4), FIG 4 (003, 4– 5)
FIG 5 (003, 6– 7)

PAR93

SEN03 1 It is perhaps cautionary to note that the equation ob-
11 tained by application of the standard statistical techniques
19 used here to the 481 data points depends on the form
SEN06 30 assumed for correlation. Thus, if we assume

$$N_{min} = CT^aD^b\Delta\rho^c\mu_c^d\mu_d^e\sigma^f\phi^g\rho_M^h \qquad (11)$$

REQU 11 (006, 5– 6)

8 application of the BMDP program 9R gives

$$N_{min} = CT^aD^b\Delta\rho^{0.43}\mu_c^{0.08}\mu_d^{0.07}\sigma^{0.01}\phi^{0.10}\rho_M^{-0.56} \qquad (12)$$

REQU 12 (006,12–13)

PAR96

SEN03 1 However, if μ_c and μ_d in eq 11 are replaced by the single
18 quantity μ_M, the result from program 9R becomes

$$N_{min} = CT^aD^b\Delta\rho^{0.41}\mu_M^{0.06}\sigma^{0.06}\phi^{0.09}\rho_M^{-0.56} \qquad (13)$$

REQU 13 (003,22–23)

PAR99

SEN03 1 The correlation coefficients for eq 12 and 13 are 0.952
SEN06 12 and 0.949, respectively. (These values exceed that for eq
[illegible] present in eq 12 and
SEN09 19 13 than the five independent group variables in eq 8.) One
3 may note the changes in exponent on σ and on ρ_M in eq
16 12 and 13 resulting from replacement of μ_c and μ_d by μ_M.

PAR102

SEN03 1 **Scale-up.** The criterion of equal power input per unit
SEN06 10 volume on large and small scales of operation has long been
21 advocated for duplicating effects on the two scales of

TXT12
PAR102
SEN09 30 mixing. This proposition is examined below for the cases
10 of full geometric similarity and of variable T/D at constant
20 W/D, H/T, B/T, and ϕ, respectively.
PAR105
SEN03 1 **(a) Full Geometric Similarity on Two Scales of**
SEN08 10 **Operation.** The present work was in the region of N_{Re}
SEN09 11 $> 10^4$, denoting turbulence throughout. In this case, with
6 full baffling, N_P is not a function of N_{Re} (Skelland, 1967).
SEN12 1 Then $P/N^3D^5\rho_M$ is constant for a given system, and since
12 V is proportional to D^3,

$$\frac{P}{V} \propto \frac{P}{D^3} \propto \frac{N^3D^5}{D^3} \tag{14}$$

REQU 14 (012,16–17)

SEN15 1 For constant physical properties, T/D, and ϕ, eq 10 shows
SEN18 12 that $N_{min} = (\text{constant})D^{-0.71}$. Substituting,

$$\frac{P}{V} \propto D^{-0.13} \tag{15}$$

REQU 15 (018, 2– 3)

PAR108
SEN03 1 Evidently the power consumption per unit volume de-
9 creases with the increasing size of the apparatus to obtain
19 the minimum impeller speed for complete dispersion.
SEN08 1 Clearly the "rule" of equal power input per unit volume
12 is in error in the safe direction.

← delete "the"

(evidently inserted by some misguided English graduat

PAR111
SEN03 1 For constant physical properties, full geometric simi-
8 larity, and constant ϕ, eq 10 gives the following for scales
19 1 and 2:

$$\frac{N_{min,1}}{N_{min,2}} = \left(\frac{D_2}{D_1}\right)^{0.71} \tag{16}$$

REQU 16 (003,22–23)

PAR114
SEN03 1 **(b) Variable T/D on Two Scales of Operation.**
SEN08 1 Equation 10 may be written in the following form for
12 constant physical properties and ϕ:

$$N_{min} = (\text{constant}')\left(\frac{T}{D}\right)^{\alpha} D^{-0.71} \tag{17}$$

REQU 17 (006,17–18)

SEN09 1 Then

$$\frac{P}{V} \propto \frac{N^3D^5}{T^6} \propto \left(\frac{T}{D}\right)^{3\alpha-2.87} T^{-0.13} \tag{18}$$

REQU 18 (009, 2– 3)

SEN12 1 Values of $3\alpha - 2.87$ for each set of data appear in Table
15 III.
PAR117
SEN03 1 Suppose that scale-up is to be accomplished in a large
SEN06 12 vessel of fixed diameter T. The negative nature of $3\alpha -$
8 2.87 in the case of single propellers and double curved-
17 blade turbines means that power input per unit volume
SEN09 26 decreases with decreasing D for these impellers. The op-
3 posite conclusion holds for the pitched-blade turbines,
10 flat-blade turbines, single-curved-blade turbines, and the
SEN12 16 disk turbine. It is therefore concluded that when using
9 pitched-blade turbines, flat-blade turbines, single-
13 curved-blade turbines, or disk turbines, it is more eco-
21 nomical to obtain complete dispersion with large impellers
SEN15 29 rotating at low speeds. The reverse is true for the other
9 impellers featured in this investigation.

9/

TXT15
SEN03 1 **Conclusions**
PAR120
SEN03 1 The present study is believed to be an advance on the
13 earlier work by Skelland and Seksaria (1978) in the fol-
22 lowing ways:
PAR123
SEN03 1 A new correlating form is used, involving the composite
11 properties ρ_M and μ_M.
PAR126
SEN03 1 Results from three different sources, van Heuven and
10 [illegible] Skelland and Seksaria (1978), and the present
19 work, are combined into an overall correlation.
PAR129
SEN03 1 This has resulted in a 2.47-fold increase in the number
12 of data points, from 195 to 481.
PAR132
SEN03 1 Six new liquid–liquid systems are introduced—four from
9 van Heuven and Beek and two from this study—to give

TXT15
PAR132
SEN06 19 a total of 11 systems. This broader base has permitted a
8 better distribution of dependencies upon physical prop-
14 erties than in the previous study.

PAR135
SEN03 1 Three new variables—H, T, and ϕ—have been included
10 in the experimental data.

PAR138
SEN03 1 Observations for a fifth type of impeller—the six-bladed
10 disk turbine used by van Heuven and Beek—are now in-
19 cluded in the data collection.

PAR141
SEN03 1 Improved statistical correlation has resulted from the
9 use of the BMDP Statistical Software program.

PAR144
SEN03 1 Nevertheless, the qualitative scale-up conclusions remain
8 essentially the same, as do the observations regarding
16 choice of impeller and its location, the types of mixing
26 phenomena that occur, and which phase becomes dis-
33 persed.

TXT18
PAR147
SEN00 1 ## Acknowledgment

SEN03 1 We are grateful to L. Forney for helpful discussion.
SEN06 1 Portions of this work were supported by National Science
11 Foundation Grants CPE 80-19617 and CPE-8203872.

TXT21
PAR150
SEN00 1 ## Nomenclature

SEN03 1 PAR153 B = baffle width, m

SEN03 1 PAR156 C, C_0, C' = constants—"shape factors"

SEN03 1 PAR159 D = impeller diameter, m

SEN03 1 PAR162 D_p = particle diameter, m

SEN03 1 PAR165 g = acceleration due to gravity, m/s^2

SEN03 1 PAR168 H = height of liquid in the vessel, m

SEN03 1 PAR171 N_{Bo} = Bond number, $D^2g\Delta\rho/\sigma$

SEN03 1 PAR174 N_{Fr} = Froude number, $DN^2\rho_M/g\Delta\rho$

SEN03 1 PAR177 N_{Ga} = Galileo number, $D^3\rho_M g\Delta\rho/\mu_M^2$

SEN03 1 N_{min} = minimum rotational speed of impeller for complete
11 liquid–liquid dispersion in agitated, baffled vessels without
16 regard to uniformity, rev/s
PAR180

SEN03 1 PAR183 P = power input to the system, W

SEN03 1 PAR186 R = weight fraction of solids

SEN03 1 PAR189 T = tank diameter, m

SEN03 1 PAR192 V = volume of total liquid, m^3

SEN03 1 PAR195 W = width of impeller blade, m

SEN03 1 PAR198 α, α_0, α' = constants

SEN03 1 PAR201 μ_c, μ_d = viscosities of continuous and disperse phases, N s/m^2

SEN03 1 PAR204 $\mu_M = \mu_c[1 + \phi(1 + 1.5\ \mu_d\phi/\mu_d + \mu_c)]$, N s/m^2

SEN03 1 PAR207 ρ_c, ρ_d = densities of continuous and disperse phases, kg/m^3

SEN03 1 PAR210 $\rho_M = \phi\rho_d + (1 - \phi)\rho_c$, kg/m^3

SEN03 1 PAR213 $\Delta\rho = |\rho_c - \rho_d|$, kg/m^3

SEN03 1 PAR216 σ = interfacial tension, N/m

SEN03 1 PAR216 ϕ = volume fraction of disperse phase

REF03
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awful (and wrong); replace with: "defined by eq 7,"

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HDG03

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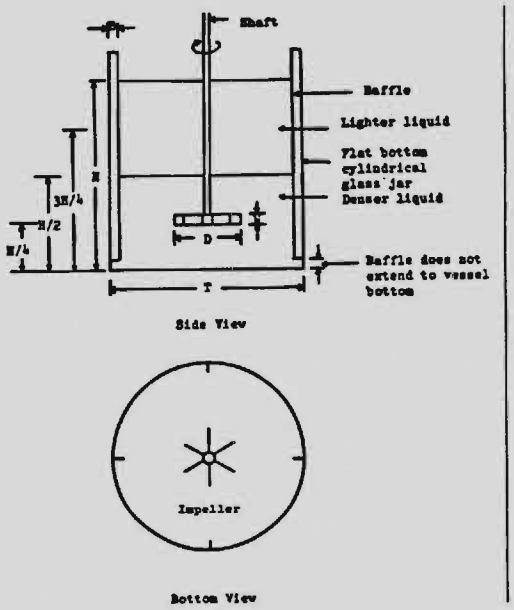
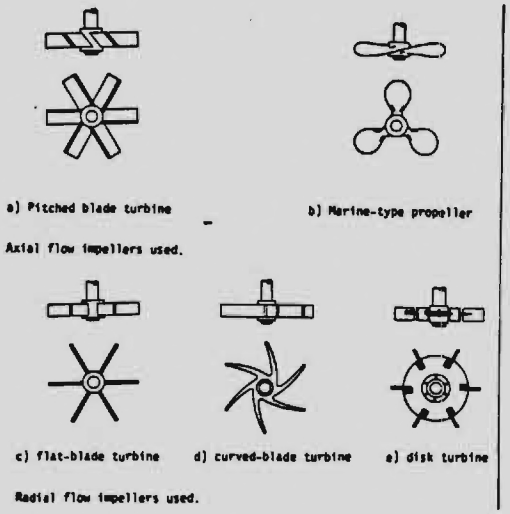
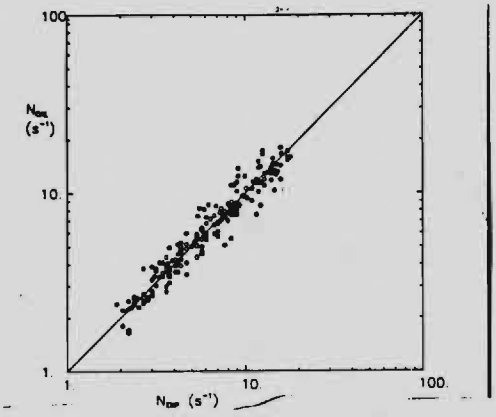


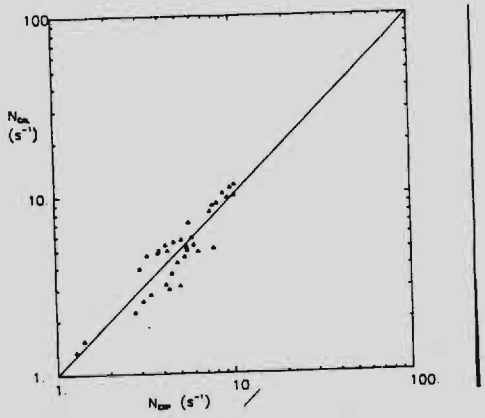
REF48
CAP00 1 **Figure 1. Schematic diagram of the experimental apparatus.**



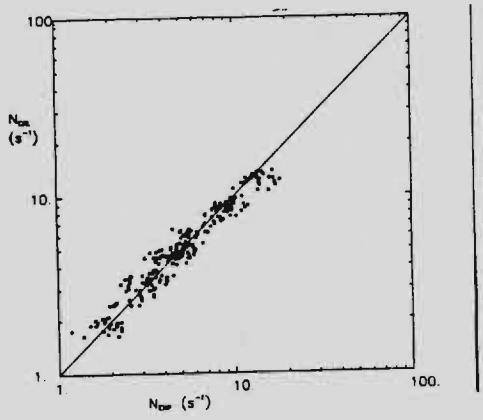
CAP00 1 **Figure 2. Five types of impellers for which eq 8-10 have been**
CAP03 12 **developed.**



CAP00 1
CAP03 13 **Figure 3.** Application of eq 8 to the published data of Skelland and Seksaria (1978).



CAP00 1
CAP03 13 **Figure 4.** Application of eq 8 to the published data of van Heuven and Beek (1971).



CAP00 1
CAP03 13 **Figure 5.** Application of eq 8 to the data obtained in the present work.

UNIT NO. 168 850805
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Ns/m²

TTL20 **Table I: Fluid Properties at 23 °C**

HDB40

	fluid	density, kg/m³	dynam viscos, N-s/m²	interfac tension with water, N/m
ROW50	ethyl acetate	894	0.000 46	0.006 27
ROW60	benzaldehyde	1041	0.001 4	0.014 5
ROW70	chlorobenzene	1106	0.001 0	0.035 2
ROW80	carbon tetrachloride	1590	0.001 0	0.045
ROW90	water	997	0.000 9	

AID00 INITIAL TABLE WIDTH IS SINGLE COLUMN

TTL20 **Table II. Apparatus Dimensions**

HDB40

	int diam of vessel, m	liq ht in vessel, m	baffle width, m	impeller diams, m
ROW50	0.216	0.216	0.019	0.102, 0.076, 0.065
ROW60	0.241	0.121	0.019	0.102, 0.076, 0.065
ROW70	0.241	0.241	0.019	0.102, 0.076, 0.065
ROW80	0.241	0.362	0.019	0.102, 0.076, 0.065
ROW90	0.292	0.292	0.025	0.102, 0.076

AID00 INITIAL TABLE WIDTH IS SINGLE COLUMN

TTL20 **Table III. Correlations and Average Deviation between $N_{min,expt}$ and $N_{min,mixd}$[a]**

HDB40

	set	H/T	impeller locat	C	α	95% confid interv on α	% av deviat	$3\alpha - 2.87$
ROW50 square pitch,	1	1	$H/4$	4.38	0.67	±0.44	16.9	−0.86
ROW60 downthrusting	2	1	$3H/4$	2.76	0.95	±0.44	14.4	−0.02
ROW70 propeller	3	1	$H/2$	4.33	0.79	±0.44	17.7	−0.50
ROW80 (three blades)	4	1	$H/4$, $3H/4$	1.46	1.33	±0.44	13.7	1.12
ROW90 downthrusting	5	1	$H/4$	1.95	1.44	±0.47	13.9	1.45
ROW100 pitched-blade	6	1	$3H/4$	1.96	1.17	±0.47	14.2	0.64
ROW110 turbine	7	1	$H/2$	0.84	1.97	±0.47	18.6	3.04
ROW120 (six blades)	8	1	$H/4$, $3H/4$	0.94	1.27	±0.47	10.0	0.94
ROW130 flat-blade	9	1	$H/4$	0.91	2.02	±0.45	9.9	3.19
ROW140 turbine	10	1	$3H/4$	*	*	*	*	*
ROW150 (six blades)	11	1	$H/2$	0.95	1.38	±0.13	11.5	1.27
ROW160	12	1	$H/4$, $3H/4$	*	*	*	*	*
ROW170	18	1/2	$H/2$	0.70	1.24	±0.28	10.2	0.85
ROW180	19	3/2	$H/2$	1.10	1.70	±0.28	14.7	2.23
ROW190 curved-blade	13	1	$H/4$	1.03	1.86	±0.46	10.2	2.71
ROW200 turbine	14	1	$3H/4$	*	*	*	*	*
ROW210 (six blades)	15	1	$H/2$	1.34	1.20	±0.46	8.9	0.73
ROW220	16	1	$H/4$, $3H/4$	1.20	0.94	±0.46	10.7	−0.05
ROW230 disk turbine	17	1	$H/2$	0.53	1.70		15.8	2.23
ROW240 (six blades)								

FNT250 [a] Asterisks indicate insufficient data due to splashing.

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Robert L. Pigford, *Editor*

March 8, 1985

Professor A. H. P. Skelland
School of Chemical Engineering
Georgia Institute of Technology
Atlanta, Georgia 30332

Dear Professor Skelland:

Here are reports from two reviewers who have studied your paper with S. Woo and G. G. Ramsay on "Effects of Surface-active Agents on Drop Size, Terminal Velocity, and Droplet Oscillation in Liquid-Liquid Systems," our manuscript number FD840-051—+. Both have recommended publication after you have revised your manuscript. I agree because your evaluation of the several previous correlations of drop properties and your new data appear to be valuable.

Reviewer I is concerned that your manuscript be clear, both in the discussion of your own experimental data and in your handling of other people's empirical formulae. His suggestions seem to me to be helpful.

I agree, also, with most of the suggestions from Reviewer II, though I don't agree that the absence of a "more satisfactory model of your own" is a cause of concern. However, I also wondered as I read your manuscript if you attempted to distinguish on statistical grounds between the random scatter of data with and without surfactants to see whether observed differences could have occurred purely through chance. I agree with this reviewer that your table I would be clearer if the surface-tension values were included in the table.

Please return a corrected paper to Washington. I plan to publish it.

Very truly yours,

Robert L. Pigford
Editor

RLP:jd
enc.
cc: Janice L. Fleming, Manager, Manuscript Office

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EFFECTS OF SURFACE-ACTIVE AGENTS ON DROP SIZE, TERMINAL VELOCITY, AND DROPLET OSCILLATION IN LIQUID-LIQUID SYSTEMS

by

A. H. P. Skelland*, Sally Woo and George G. Ramsay
School of Chemical Engineering
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Atlanta, Georgia 30332

ABSTRACT

Drop size, terminal velocity, and the onset of oscillation of chlorobenzene drops falling through water were determined in systems containing anionic, cationic, or nonionic surfactants. Surfactant concentration, nozzle diameter, and nozzle velocity were varied to cover the range of most industrial applications of drop formation from circular orifices in the nonjetting region.

Existing correlations for drop size and terminal velocity in uncontaminated systems are shown to be adequate in the presence of surfactants (SAA) when used with the diminished value of interfacial tension due to the SAA. The applicability of criteria for the onset of oscillation in pure systems to those containing surfactants is examined.

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Ms. No. 4697 **Title:** The Effects of Surface Active Agents on Jet Breakup in Liquid-Liquid Systems

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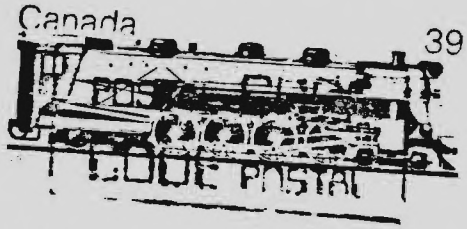

Dr. A.H.P. Skelland
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ATLANTA, GEORGIA
30332-0100 USA

sent Jan 12, '87

THE EFFECTS OF SURFACE ACTIVE AGENTS ON JET BREAKUP IN LIQUID-LIQUID SYSTEMS

By

A. H. P. Skelland and Perry G. Walker*

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The Georgia Institute of Technology
Atlanta, GA 30332

*Perry G. Walker is now with E. I. du Pont de Nemours at the Savannah River Plant, S.C.

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January 12, 1987

Dr. N. Epstein, Editor
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Canada

Dear Dr. Epstein:

I enclose three copies of the paper on "The Effects of Surface Active Agents on Jet Breakup in Liquid-Liquid Systems," by myself and Mr. Perry G. Walker, which you may consider suitable for publication in the Canadian Journal of Chemical Engineering.

Yours sincerely,

A. H. P. Skelland
Professor of
Chemical Engineering

AHPS/jvl

ABSTRACT

An experimental study has been made of the effects of anionic, cationic, and nonionic surface active agents on jet length, jet contraction, and drop size resulting from the formation and breakup of liquid jets in another immiscible liquid, while undergoing mass transfer.

Correlations previously established for uncontaminated systems with various rates of mass transfer successfully fitted the present data, when used with the diminished value of interfacial tension caused by the surfactant. Variables included nozzle size, flow rate, and type and concentration of surfactant. Data were obtained from measurements on over 3,000 drops, plus jet lengths and jet diameters from 501 photographs.

Conclusions

New data are presented on the effects of anionic, cationic, and nonionic surface active agents on jet length, jet contraction, and drop size in a liquid-liquid system undergoing moderate rates of mass transfer.

Existing correlations, obtained from uncontaminated systems with "moderate" rates of mass transfer, were found to correlate the new data with S.A.A. for jet length and jet contraction within the rather broad limits of accuracy to be expected for these fluctuating quantities.

In contrast, drop size from jet break-up was best correlated by a relationship previously obtained for S.A.A.-free sytems with "very low" rates of mass transfer.

All correlations were applied using the diminished value of interfacial tension caused by the presence of the surfactant.

Transfer was from the disperse to the continuous phase throughout. No distinctions were found between the effects obtained from the three different surfactants used on L_j, d_{jc}, and d_D at a given value of interfacial tension.

Acknowledgement

This work was partially supported by National Science Foundation Grants numbered CPE82-03872 and CPE82-03872/01.

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Donald R. Paul, Editor

Author(s) A. H. P. Skelland, L. T. Moeti

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Ms No.

Ms Title Effects of Surface Active Agents on Minimum Impeller Speeds for Liquid-Liquid Dispersion in Baffled Vessels

January 16, 1987
Received

This manuscript is being considered with the understanding it has been submitted on an exclusive basis. The office noted below will write you of our decision regarding it as soon as possible.

A. H. P. Skelland
School of Chemical Engineering
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Sent Jan 13, '87

EFFECTS OF SURFACE ACTIVE AGENTS ON MINIMUM IMPELLER SPEEDS FOR LIQUID-LIQUID DISPERSION IN BAFFLED VESSELS

by

A. H. P. Skelland and L. T. Moeti

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Dear Sir:

I enclose four copies of the paper on "Effects of Surface Active Agents on Minimum Impeller Speeds for Liquid-Liquid Dispersion" by myself and Dr. L. T. Moeti which you may consider suitable for publication in the new journal "Industrial and Engineering Chemistry Research".

Yours sincerely,

A. H. P. Skelland
Professor of
Chemical Engineering

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ABSTRACT

The effects have been examined of nonionic, anionic, and cationic surface active agents (S.A.A.) on the minimum impeller speed for complete dispersion in agitated vessels. Variables included size and form of impeller, fluid properties in two equal-volume liquid-liquid systems, and surfactant concentration. The Skelland and Ramsay (1983) correlation

$$\frac{N_{min}^2 \rho_M D}{g\Delta\rho} = C^2 \left(\frac{T}{D}\right)^{2\alpha} \phi^{0.106} \left(\frac{\mu_M^2 \sigma}{D^5 \rho_M g^2 \Delta\rho^2}\right)^{0.084}$$

served to predict the minimum impeller speed, when used with diminished interfacial tensions due to the presence of S.A.A., with an overall absolute deviation of 11.67%. This contrasts with some droplet phenomena, where more elaborate allowance for contamination effects are required (Mekasut et al., 1979). The difference between deviations among the surfactant systems was accounted for by experimental error and verified by two methods of statistical analysis.

particular species of surface active contaminants used here. Rather, the surfactants are intended to simulate some of the impurities encountered industrially, which are usually unknown with regard to number of species, structure, and concentration. The object, then, is to ascertain whether characterization of their effects by measurement of a single property, σ, is adequate for purposes of prediction of N_{min} via equation 10 without further modification. The indications from this study are in the affirmative.

For developments regarding scale-up the reader is referred to Skelland and Ramsay (1983).

ACKNOWLEDGEMENTS

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January 12, 1987
Received

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EFFECTS OF SURFACE ACTIVE AGENTS ON DROP SIZE IN LIQUID-LIQUID SYSTEMS

By

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I enclose four copies of the paper on "Effects of Surface Active Agents on Drop Size in Liquid-Liquid Systems" by myself and Mrs. Elizabeth A. Slaymaker, which you may consider suitable for publication in the new journal "Industrial and Engineering Chemistry Research".

Yours sincerely,

A. H. P. Skelland
Professor of
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ABSTRACT

Extensive new experimental data are presented on the effects of anionic, cationic, and nonionic surface active agents on the size of drops formed at a nozzle tip in liquid-liquid systems.

The Scheele and Meister equation for predicting drop size in surfactant-free systems was found to give generally satisfactory agreement with these data, when used with the diminished equilibrium value of interfacial tension caused by the surfactant, for drop formation times of 0.5 to 4 seconds. Various minor trends are identified within this general area of agreement.

The need to confine use of the Scheele and Meister equation to its specified range of nozzle diameters is confirmed by the present data, in both the presence and the absence of surface active agents.

equilibrium value of interfacial tension due to the surfactant, for drop formation times of 0.5 to 4 seconds.

Thus, the mean deviation between calculated and experimental drop volumes in the presence of surfactants was 1.8% and the mean absolute deviation was 12.9%. For our systems without added surfactant, the mean deviation was 6.8% and the mean absolute deviation was 9.5%. Various minor trends are identified within this general area of agreement.

The importance of applying the Scheele and Meister equation only within its specified range of nozzle diameters is confirmed by the present data, obtained both with and without surface active agents.

ACKNOWLEDGEMENT

This work was partially supported by National Science Foundation Grants numbered CPE82-03872 and CPE82-03872/01.

DISPERSE-PHASE MASS TRANSFER IN AGITATED LIQUID-LIQUID SYSTEMS

By

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*Hu Xien is at Tsinghua University, Beijing, P.R.C.

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ABSTRACT

Disperse-phase mass transfer rates were measured in a baffled vessel agitated with 6-flat-blade turbines. The results from three liquid-liquid systems were used to select the better of two theoretical models of the process and were also correlated with operating variables and physical properties. The internal state of the drops is considered.

values predicted by equation (38) was 9.8%. Figure 5 shows a plot of predicted versus experimental k_d for the 55 runs correlated.

Acknowledgements

This work was partially supported by the National Science Foundation Grants CPE-8203872 and CPE-8203872/01. Hu Xien was supported by Tsinghua University, Beijing, P.R.C.

DROP BREAKAGE IN AGITATED LIQUID-LIQUID SYSTEMS

By

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ABSTRACT

An expression is developed for the drop breakage frequency in liquid-liquid systems agitated by a 6-flat-blade turbine in a baffled vessel. This is based on the OPED model of Molag et al (1980). The resulting relationship contains only one constant, k, which is evaluated from experiments on three liquid-liquid systems and correlated with operating conditions and physical properties by dimensional analysis.

Values of Sauter mean drop diameter at various times during each run were calculated from the correlation for comparison with measured values; the mean absolute deviation for 75 data points was 10.5%.

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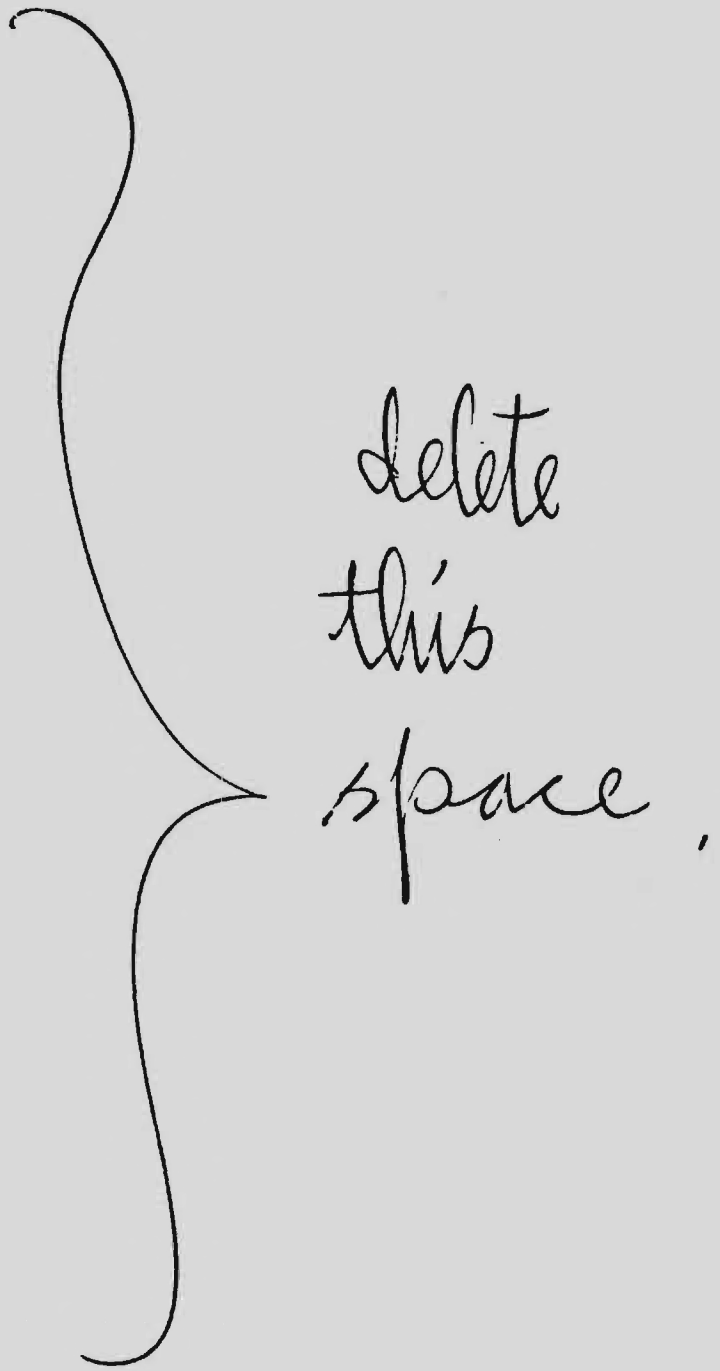
This work was partially supported by the National Science Foundation Grants CPE-8203872 and CPE-8203872/01. Hu Xien was supported by Tsinghua University, Beijing, P.R.C.



c) Scientific Collaborators

1) A. H. P. Skelland, Professor and Principal Investigator.

2) Hu Xien, Visiting Scholar, a faculty member of Tsinghua University, Beijing, PRC.

3) Nicholas C. Vasti, graduate student.

4) George G. Ramsay, graduate student.

5) Sally Woo, graduate student.

6) Perry G. Walker, graduate student.

7) Lebone T. Moeti, graduate student.

8) Elizabeth A. Slaymaker, graduate student.

e) Technical Description of Project and Results

This report describes work performed in the author's laboratory - partly with the support of NSF Grant No. CPE-8203872 - on liquid-liquid dispersion and mass transfer in agitated, baffled vessels, as used in liquid-liquid extraction, batch polymerization processes, and chemical reactors.

Part I. Minimum Impeller Speeds for Newtonian Liquid-Liquid Dispersion in Baffled Vessels.

With the single exception of the paper by van Heuven & Beek (1971), the only Newtonian fluid studies in this area have been those by Skelland and Sek saria (1978), Skelland and Lee (1978), Skelland and Ramsay (1983) and (1987), and Skelland and Moeti (1984).

The latest and most comprehensive study of the minimum impeller speed required for complete dispersion (Skelland and Ramsay, 1987) was based on variables which included five common types of impeller, two generating axial flow and three radial flow, in four locations; fluid properties in eleven systems; tank diameter (T); liquid height (H); and volume fraction of the disperse phase (ϕ). Observations from 481 runs drawn from three different sources were correlated with an average absolute deviation of 12.7% by the expression

$$(N_{Fr})_{min} = C^2\left(\frac{T}{D}\right)^{2\alpha}\phi^{0.106}(N_{Ga}N_{Bo})^{-0.084}$$

where N_{fr}, N_{Ga}, and N_{Bo} are the Froude, Galileo, and Bond numbers, D is impeller diameter, and C and α are tabulated according to impeller type, location, and - in some cases - the ratio H/T. Scale-up relationships were developed from the above expression. The paper is currently in the galley

proof stage of publication in Industrial and Engineering Chemistry Research.

The paper with L. Moeti (1984) shows experimentally that the above equation is adequate in the presence of surface active contamination (which commonly occurs industrially), provided that it is used with the diminished value of interfacial tension (in N_{Bo}) due to the presence of the surfactant. The paper was first presented in the symposium on "Interfacial Phenomena in Liquid Extraction," chaired by Prof. W. J. Heideger, at the San Francisco meeting of the AIChE in November, 1984, and is now in the course of publication.

The above studies, first outlined in the proposal leading to NSF Grant No. CPE-8203872, are now complete. Shortage of NSF funds led to provision of only part of the support requested, preventing study of the effects of making first the continuous phase, then the disperse phase, and finally both phases non-Newtonian, by the addition of such materials as Carbopol 934 in various small concentrations.

Part II Mass Transfer in Agitated Liquid-Liquid Systems

L. T. Moeti performed 180 runs in agitated liquid-liquid systems in baffled vessels, to measure the continuous phase mass transfer coefficient in high and low interfacial tension systems. He found that

$$k_c a \propto N^{1.5} \mu_c^{-1/3} D^{2/3}$$

where N is impeller speed, μ_c is the continuous phase viscosity, and D is solute diffusivity in the continuous phase. Agitation was provided by a centrally mounted turbine with six flat blades.

Hu Xien, a one-year visiting scholar from Tsing Hua University in

Beijing, P.R.C., performed 86 runs on three agitated liquid-liquid systems, to measure the disperse phase mass transfer coefficient for the first time in such equipment. His results were used to select the better of two highly preliminary models of the process and were also correlated empirically in dimensionless form. The enclosed papers outline the work more fully.

Drop sizes in both studies were known from measurement of photographs taken during each run.

The merits of batch operation in elucidating transfer mechanisms and in evaluating k_d and k_c have not perhaps been fully appreciated. Thus droplet breakdown into successively smaller drops persists for several minutes after injection of the disperse phase (Skelland and Lee, 1981; Bapat et al, 1983; Bapat and Tavlarides, 1985; Hong and Lee, 1985), whereas mass transfer is usually complete after a minute or two (Rushton et al, 1964; Treybal, 1980; Skelland and Lee, 1981). Batch operation in the manner used here enables this behavior to be quantitatively followed with time by measurement of k, drop size, and surface area from moment to moment for a single injected element of disperse phase. In contrast, continuous operation gives only average transfer conditions corresponding to many different elements of disperse phase which simultaneously occupy the vessel but which entered it at different times and which are therefore in different stages of mass transfer and drop breakup at any given instant. The time history of a given element of disperse phase in the vessel cannot then be followed in the manner necessary to properly model the process.

Part III <u>Additional accomplishments that have been partially aided by NSF Grant No. CPE-8203872</u>

The following four sets of research results, although not obtained in

agitated vessels, show closely related aspects of droplet behavior in liquid-liquid systems. They were all performed while the author and G. G. Ramsay were partly supported by NSF Grant No. CPE-8203872.

The paper by Skelland and Vasti deals with the effects of interaction between droplets in swarms on various transport processes. The paper by Skelland, Woo, and Ramsay shows the effects of surface active contamination on some aspects of drop behavior, including oscillation; the enclosed letter from Dr. R. L. Pigford, editor of I/EC Fundamentals, states that he intends to publish it after revision. Also included are papers, title pages, and abstracts of the M.S. theses of P.G. Walker and Elizabeth Slaymaker, on the effects of surfactants on jet breakup and drop size in liquid-liquid systems. These studies are now being reviewed for publication.

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